

**USES OF
ADVANCED PULSED NEUTRON SOURCES**

**Report of a Workshop Held at
Argonne National Laboratory
October 21-24, 1975**



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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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USES OF
ADVANCED PULSED NEUTRON SOURCES
Report of a Workshop Held at
Argonne National Laboratory
October 21-24, 1975

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ABSTRACT

This report contains the conclusions that were drawn by nine panels of scientists in the fields of Biology; Chemical Spectroscopy; Chemical Structures of Crystalline Solids; Chemical Structures of Disordered Solids and Inhomogeneous Systems; Dynamics of Solids Liquids, Glasses and Gases; Magnetism; Neutron Sources; and Radiation Effects. The nine panel reports describe the applications found in these scientific areas, accompanying them with conceptual instruments designed for the measurements and with calculations to establish feasibility.

EDITORS' PREFACE

This report contains the conclusions that were drawn by the nine Panels of scientists who met at Argonne for a Workshop on Uses of Advanced Pulsed Neutron Sources. The individual Panel reports were completed in draft form and distributed to Workshop attendees on the last day of the meetings. Each Panel chairman, with input from those in his Panel, has reviewed and provided whatever corrections were required to the contents of the drafts. We have done some minor editing as well, mostly to put them into consistent format. The participants in each Panel and their affiliations are listed at the beginning of each Report, and also in a separate list. They should be considered the co-authors of their Reports with responsibility only for the contents of their Panel Report.

The Conference and Workshop, and this Report, represent a significant event in the history of the uses of neutrons for condensed matter research. Here a group of experts set down its statement of where the science stands, and looking ahead to the advent of advanced pulsed neutron sources with unique new performance, projected their assessment of where the science will be and what it should be. We have attempted to summarize the most exciting prospects for the uses of advanced pulsed neutron sources below in a separate section, "Summary of Highlights."

The Workshop Panel members were charged with two tasks--to compile scientific problems, and to propose and evaluate the instrumental means to accomplish the measurements. Thus the Panel Reports contain not only discussions of scientific applications opened up by advanced pulsed neutron sources, but also descriptions of proposed instruments and evaluations of their performance. A number of brand new instrument ideas were generated which are described in the Reports. The Statement of Goals presented to the participants is included below.

The Workshop participants were provided copies of the Draft Proposal for an Intense Pulsed Neutron Source (which will shortly appear in final form) in which data are given for purposes of slow neutron pulse width and intensity calculations, and for evaluation of fast neutron fluxes in the IPNS radiation effects facility. Conceptual designs of several instruments devised for IPNS were also provided to the participants.

The Conference and Workshop took place October 20-29, 1975. At the Conference held October 20, ten speakers reviewed the uses of neutrons in various areas and the developing and proposed advanced pulsed neutron sources. The rest of the week was devoted to the Workshop, in which the Panels met individually to develop their conclusions. The program for the Conference and Workshop is included below.

On June 30 and July 1, 1975, a planning group consisting of M. Hamermesh, University of Minnesota; S. W. Peterson, Argonne National Laboratory; M. T. Robinson, Oak Ridge National Laboratory; B. P. Schoenborn, Brookhaven National Laboratory; C. G. Shull, Massachusetts Institute of Technology; G. Stucky, University of Illinois, Urbana; and J. M. Carpenter, Argonne National Laboratory, met to plan the Conference and Workshop. We acknowledge the help of the planning group in setting up such an effective program.

Further details of the Conference and Workshop were completed by the Argonne coordinators of the Workshop Panels, who were aided by Dr. Lester Guttman with further help from the Argonne pulsed neutron source users, all of whom we thank for their assistance.

The Conference and Workshop were sponsored jointly by Argonne National Laboratory and the Argonne Universities Association. The University of Chicago and AUA sponsored an informal reception and the Workshop Banquest. We are grateful for this support, which made the proceedings possible.

Mrs. Carol Oleferchik and Miss Florence Belcher prepared the final typed version of this report, and Mr. Harry Cern and Mr. Robert Stefiuk redrew the draft figures into the form in which they appear here. We thank them for their help in enabling the timely completion of this report.

Mrs. Miriam Holden and Mrs. Jean Matheson of the Argonne Conference Planning and Management office and Mrs. Linda Evans and Mr. Willis Denekas very effectively arranged the housing, transportation, and other details of the care and feeding of the attendees. We express thanks to them for all of us.

One of us (JMC) thanks the other (SAW) for providing the "little blue cards" containing numerical data on The Neutron, which were distributed at the Conference and Workshop. These very useful data, based on the 1973 tabulation of physical constants, are reproduced as a tear-out card just inside the back cover of this report.

J. M. Carpenter

S. A. Werner

BACKGROUND FOR THE CONFERENCE AND WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

The Conference and Workshop on Uses of Advanced Pulsed Neutron Sources took place at a time when a number of examples of operating pulsed neutron sources and a number of studies of advanced pulsed source systems were emerging. These include the present and the scheduled new Harwell electron linac sources in the U.K., the series of Soviet pulsed fast reactors IBR-I, IBR-30, IBR-II, the Soviet electron linac source at Kurchatov, the Japanese electron linac source operating at Tohoku, the proposed Japan Linac Booster, the KENS spallation source at Tsukuba, Japan, the operating electron linac source at Toronto, Canada, the studied application of the Oak Ridge Electron Linear Accelerator for neutron scattering, the nearly completed Weapons Neutron Research Facility at LAMPF, and the Argonne pulsed neutron sources, ZING-P, ZING-P', PPNS, IPNS I and IPNS II. The table below describes Argonne's programed sequence of prototype sources, and the final Intense Pulsed Neutron Source.

These systems are the source designers' response to the fact that the conventional neutron sources, steady state research reactors, have reached limitations imposed by heat transfer and cost, and thus the available neutron fluxes have increased by only about a factor of three in the past twenty years. The High Flux Reactor at the Institut Laue-Langevin, Grenoble, represents the current highest evolution of the steady state research reactor and provides a thermal neutron flux of about 1.2×10^{15} n/cm²-sec.

This is important because in almost all areas of neutron scattering research the source flux limits what can be done. The limitation is so severe that a great many important measurements cannot even be attempted and no realistic amount of additional experimenters' patience or instrumental cleverness can open up many new areas of research.

Pulsed neutron sources, with which time of flight measurement techniques are used, offer the means for obtaining higher useful neutron fluxes than the conventional steady state reactor sources. This is because the source is "on" for only a very small fraction of the time, and the heat which must be dissipated to produce neutrons is averaged out over all time, therefore very high instantaneous power densities and instantaneous neutron fluxes can be achieved. The short pulse provides a beam of neutrons which is already prepared for energy analysis by time of flight, and the entire spectrum can be used for measurement. By contrast, when neutron beams from steady reactors are used, a very large fraction (~99%) of the neutrons are thrown away in preparing the beams. The differences in the ways pulsed and steady beams are used make direct comparisons of pulsed and steady sources difficult. Only by comparing data rates in instruments designed for the same purpose on the two types of sources, can a really valid comparison be made. However, the peak flux in a pulsed source can be very roughly compared with the steady flux in a steady source to evaluate their relative value if the pulsed source repetition frequency is on the order of 100 Hz. On this basis, the pulsed source is usually underestimated relative to its evaluation on the basis of instrument data rates.

ANL Pulsed Neutron Source Program

<u>Source Neutron</u>	<u>Proton Accelerator</u>	<u>Protons/ Pulse</u>	<u>Proton Energy (MeV) and Target</u>	<u>Neutrons/ Proton</u>	<u>Frequency[†] c/sec</u>	<u>Peak Thermal Neutron Flux (n/cm² sec)</u>
ZING-P	ZGS Booster I	2.5×10^{10}	200 Pb	2	10	5×10^{11}
ZING-P'	ZGS Booster II	1×10^{12}	500 W	8	$2/3 \times 30$	1×10^{14}
PPNS	ZGS Booster II	$1-5 \times 10^{12}$	500 W	8	$.9 \times 30$	$1-5 \times 10^{14}$
IPNS Phase I	ZGS Booster II	5×10^{12}	500 ^{238}U	20	$.9 \times 30$	10^{15}
IPNS Phase II	HIS [*]	5×10^{13}	800 ^{238}U	30	60	10^{16}

^{*} High Intensity Synchrotron

[†] The first factor is the expected fraction of time available for neutron production, the second factor is the repetition frequency.

The pulsed sources provide an advantage over steady reactors in the neutron energy range above the thermal neutron energies. This is because the moderators used are small and of hydrogen while large D₂O or Be moderators are the choice in steady reactors. The hydrogenous moderators provide short pulses of slow neutrons, but are relatively inefficient. Thus, a pulsed source which provides a certain thermal neutron flux, provides a much larger peak epithermal neutron flux than the steady reactor which has a thermal flux equal to the pulsed source peak flux. The differences are factors of the order of 100 at 1. eV. It is to overcome this deficiency that the 2000°K Hot Source has been installed at the ILL High Flux Reactor. This very high epithermal neutron flux, accompanied by very short pulse widths in the epithermal range, is a unique new feature of the pulsed neutron sources, which has been foreseen in the Panel Reports to open up many new important possibilities for neutron scattering research.

The spallation neutron sources have a characteristic feature which makes them attractive for radiation effects studies, in which the pulsed operation has been found neither to be disadvantageous nor greatly advantageous. This feature is that the spallation mechanism of neutron production leads to release of relatively few gamma rays per neutron, by comparison with the fission sources which have commonly been used for this research. An intense photon flux heats samples under irradiation. In the many cases where careful temperature control or very low temperatures are required, the unique low photon flux accompanying neutron production in a spallation source makes possible measurements which would otherwise be unfeasible. This, with the high time average fluxes that can be produced, makes the spallation sources exceedingly attractive for neutron radiation effect research.

These considerations led to the convening of the Conference and Workshop on Uses of Advanced Pulsed Neutron Sources.

BRIEF SUMMARY OF HIGHLIGHTS OF THE WORKSHOP

It has been recognized for some time that thermal neutron scattering is the most general experimental technique in condensed matter research. In many cases it yields direct microscopic information which is inaccessible by any other technique, and in many other cases produces results which are completely complementary to the information obtained from other physical measurement methods. Applications spanning biology, chemistry, physics and materials science are well established and are growing constantly.

It is clear from the panel reports of this workshop that the construction of an advanced pulsed neutron source, such as IPNS, would expand the realm of scientific problems accessible to neutron scattering enormously in many directions. The reason for this is attributable to four general characteristics of such a source;

- Very high intensity throughout the thermal energy range.
- High epithermal flux (λ 0.1 eV)
- Pulsed nature of the source
- The moderator (and therefore the flux spectrum and pulse widths) can be easily tailored to meet specific needs.

The region of energy and wavevector transfers in current neutron scattering experiments, using thermal reactors is, roughly speaking below 150 meV and 10 \AA^{-1} . Experiments of fundamental and practical importance were proposed in this workshop which require energy transfers up to about 10 eV, and in other cases wavevector transfers out beyond 50 \AA^{-1} . Some of these are feasible with an advanced pulsed neutron source.

Due to the pulsed nature of the source, very high pulsed magnetic fields, pulsed electric fields, or pulsed pressures can be applied to a sample. Certain time-dependent phenomena having relaxation times in the millisecond to minute range can be investigated utilizing the pulsed nature of the incident flux. Since the source is "off" between pulses, the background is lower in a pulsed neutron source than in an equivalent steady source.

A very high incident intensity allows experiments to be done on much smaller samples. Thus, experiments could be done on materials which are very rare, very expensive, or are small fragments of other materials.

Because an advanced pulsed neutron source would have several moderators, and each can be tailored separately to meet a specific need (and perhaps even changed from time to time), a prolific supply of both cold neutrons and epithermal neutrons can be obtained.

In short, the realm in which the neutron scattering technique is applicable could be expanded by a larger factor in many directions utilizing an intense pulsed neutron source.

In Biology the major gains would be attributable to the higher source flux. The importance of hydrogen in the dynamics and structure of living things is the key reason why the neutron scattering technique is of growing relevance in biology. Differential labelling using the difference in scattering lengths of H and D is of prime importance. To fully utilize the ideas of isotopic substitution and the anomalous scattering lengths of certain isotope such as CD^{113} and Sm^{149} , higher energy, epithermal neutrons, are required. It is extremely important that neutrons are a "gentle" form of radiation, not disrupting certain biological structures during measurement, as is the case with x-rays and electrons.

In Chemical Spectroscopy, especially in experiments on molecular excitations in fluids, it is of great importance to have a good supply of epithermal neutrons so that measurements can be done at low momentum transfers, thus reducing recoil broadening. In order to study the dynamics and structures of gases adsorbed on the surfaces of solids, catalysis, small particles, diffusion of fluids through membranes and the initial stages of intercalation, the higher incident fluxes provided by advanced pulsed neutron sources are important. The determination of the detailed shapes of molecules require data to be obtained out to high wavevectors, perhaps in some cases greater than 30 \AA^{-1} . The investigation of certain electronic transitions such as many crystal field transitions or singlet-triplet excitations, require the epithermal flux provided by IPNS. To fully utilize the polarization analysis techniques to separate incoherent and coherent scattering requires the highest possible source intensities.

The substitution of time resolution in an experiment on a pulsed machine for space resolution in experiments on steady state sources facilitates the installation of special equipment such as high pressure cells, bulky magnets and small-window low- and high-temperature equipment. This is particularly important in Chemical Structures work. The importance of the neutron technique in the study of the one-dimensional conductors KCP and TTF-TCNQ has already been demonstrated. Greater resolution, and therefore higher incident intensities, are necessary to resolve all the details of these materials. A variety of alloys and metals (such as FeTi) have been tested as possible hydrogen-storage media. Since neutrons "see" the hydrogen in these materials, basic and practical information using neutron scattering has, and will continue to be gained. The high resolution-small sample diffractometer described by the Chemical Structures Panel would provide high data collection rates on samples as small as 0.1 milligram. In order to study the details of short range order, atomic displacements, local distortions and clustering in alloys, very high resolution near Bragg peaks is necessary. To gain this resolution a very intense source is necessary.

The Dynamics of certain Solids having high $1/v$ absorption cross sections could be studied with advanced pulsed neutron sources, taking advantage of the larger epithermal flux. Applications to the study of high-lying optic modes in solids, electronic level spectroscopy, and the ground-state momentum distribution in liquids all require this large epithermal flux. The measurement of phonon dispersion, and the phonon density of states of complicated materials is very amenable to the neutron time-of-flight technique. This is particularly important for a wide variety of superconducting alloys and compounds. Of course, the time-of-flight technique is also the method best

suited for the study of $S(Q, \omega)$ in amorphous materials (glasses) and in liquids. Chalcogenide glasses and semiconductors (As, Se, Ge, Te compounds) have considerable technological applications. Their structures need to be understood in order to help clarify their electronic properties. Interest in molecular liquids, molten salts, liquid metals, electrolytes, and the various types of diffusion in liquids and solids continues to increase. The pulsed neutron source is ideally suited to study these problems.

Because of the rapid fall-off of the magnetic form factor with increasing wavevector for most materials of interest in Magnetism, the range of scattering vectors of interest is usually $Q \lesssim 4 \text{ \AA}^{-1}$. Thus, in order to complete the scattering triangle, one needs a rather high incident neutron energy to observe excitations of even rather modest energy $\hbar\omega$, say 100 meV. In order to obtain a reasonably complete measurement of the generalized susceptibility, of, say, the transition metals Cr, Fe and Ni, a large epithermal flux of neutrons is necessary. It is clear that neutron polarizing filters should be used at advanced pulsed neutron sources to produce or analyze polyenergetic beams. Appropriate filters have already been successfully tested and development is continuing. Inelastic polarization analysis experiments to obtain direct information on the relative magnitudes of magnetic and nuclear scattering, to separate the parallel and perpendicular parts of $\chi(\vec{Q}, \omega)$, and to separate spin incoherent and coherent scattering are very desirable in the study of magnetic materials. This, however, requires the highest possible fluxes. The study of samples in the presence of very large pulsed magnetic fields, and of various time-dependent magnetic phenomena are well-suited to a pulsed source.

The Radiation Effects Panel found that a fast neutron irradiation facility such as proposed for IPNS would be an attractive facility for basic radiation damage studies. Features of particular advantage are the low rate of gamma heating accompanying a useful neutron level, the variation of flux spectrum in different irradiation positions, the large available experimental volume, the possibility for some experiments of using the pulsed nature of the source, and the fact that the facility would be dedicated to use for these measurements and have close access to unique diagnostic tools such as the IPNS neutron scattering facility. The low gamma heating rate makes possible well controlled experiments at temperatures as low as 4 K, in a flux 20 times greater than now available. The flux pinning effect in Type II superconductors can be extended to higher fluences than ever possible. Careful damage rate measurements can be made at temperatures where damage production and annealing compete, and studies in situ of mechanical properties influenced by radiation produced defects can be performed.

The Neutron Sources Panel found after review of various aspects of pulsed neutron source design that advanced pulsed neutron sources at the level of IPNS performance are feasible. Heat transfer, cyclic thermal stress, thermal elastic shock stress, and radiation damage problems in the targets are not considered to be severe, although these probably require further study. The panel examined the availability of data on neutron yields from spallation targets, target-moderator coupling, and slow neutron spectra and pulse widths from moderators. Methods of calculation exist for the first two of these, and the need was recognized for further investigations in all three of

of these areas. The growing body of information is adequate for the present, but further information should be developed for detailed design and optimization of moderators for specific purposes. Monte Carlo calculations are implied here, although experimental methods may be more practical for determining slow neutron performance of moderators. Recognizing the need for these further data, the panel recommended the formation of a data sharing pool representing at least the seven pulsed neutron source efforts worldwide.

The workshop panels made preliminary designs of various neutron spectrometers and assessed their performance (numerically in most cases) in carrying out certain classes of experiments alluded to here. We list the instruments discussed here:

1. Time of flight single crystal diffractometer
2. Small angle scattering spectrometer
3. High Intensity Powder Diffractometer
4. Constant - \vec{Q} spectrometer
5. Small \vec{K} , high $\hbar\omega$ spectrometer
6. General-purpose TOF spectrometer
7. High resolution time of flight spectrometer
8. $S(Q,0)$ elastic scattering diffractometer

This summary is provided simply to provide a birds-eye view of this workshop report. It would be a great mistake on the part of any reader to stop here.

GOALS OF THE CONFERENCE AND WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

To acquaint the scientific community with the potential uses of advanced pulsed neutron sources in various fields and with examples of such sources currently under development.

To explore areas of science where advanced pulsed neutron sources will provide capability for new developments, especially in areas beyond those where neutrons are now commonly used.

To evaluate the experimental requirements for pulsed neutron sources and propose instruments and their applications.

To recommend policies for operation of a large scale pulsed neutron source.

To generate a report describing applications found, focussing on novel, imaginative and interdisciplinary applications, providing supplementary calculations which demonstrate the feasibility of these applications, and indicating what features of instruments and of sources are needed.

LIST OF PANELS AND PARTICIPANTS

Conference and Workshop on Uses of Advanced Pulsed Neutron Sources, October 20-24, 1975

I. Biology

D. L. D. Caspar*	Brandeis University
S. W. Peterson [†]	Argonne National Laboratory
G. M. Brown	Oak Ridge National Laboratory
S. S. Danyluk	Argonne National Laboratory
M. E. Druyan	Hines Veteran Hospital
B. Jacrot	Institut Laue-Langevin, France
A. C. Nunes	Institut Laue-Langevin, France
A. M. Scanu	The University of Chicago
M. Schiffer	Argonne National Laboratory
B. P. Schoenborn	Brookhaven National Laboratory

II. Chemical Spectroscopy

J. J. White*	Institut Laue-Langevin, France
J. M. Williams [†]	Argonne National Laboratory
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C. V. Berney	Massachusetts Institute of Technology
S. H. Chen	Massachusetts Institute of Technology
H. L. McMurry	University of Missouri-Columbia
J. McTague	University of California, Los Angeles
J. J. Rush	National Bureau of Standards
G. C. Stirling	Rutherford Laboratory, U.K.
H. Taub	University of Missouri-Columbia
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* Chairman

[†] Argonne Coordinator

LIST OF PANELS AND PARTICIPANTS

III. Chemical Structures of Crystalline Solids

P. Coopens*	State University of New York-Buffalo
M. H. Mueller†	Argonne National Laboratory
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J. A. Jacobson	Ames Laboratory
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M. Lehmann	Institut Laue-Langevin, France
R. B. VonDreele	Arizona State University
T. G. Worlton	Argonne National Laboratory

IV. Chemical Structures of Disordered Solids and Inhomogeneous Systems

J. B. Cohen*	Northwestern University
J. Faber†	Argonne National Laboratory
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G. Bauer	KFA Jülich, Germany
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G. Summerfield	University of Michigan

* Chairman

† Argonne Coordinator

LIST OF PANELS AND PARTICIPANTS

V. Dynamics of Solids

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D. Moncton	Bell Laboratories
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C. A. Pelizzari	Argonne National Laboratory
J. M. Rowe	National Bureau of Standards
C. W. Tompson	University of Missouri-Columbia
T. Wolfram	University of Missouri-Columbia

VI. Liquids, Glasses and Gases

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K. Carneiro	Brookhaven National Laboratory
J. E. Danner	University of Missouri-Columbia
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G. F. Mazendo	The University of Chicago
A. H. Narten	Oak Ridge National Laboratory
T. A. Postol	Argonne National Laboratory
Y. P. Sharma	University of Missouri-Columbia
K. S. Singwi	Northwestern University
K. Sköld	Argonne National Laboratory
A. D. B. Woods	Atomic Energy of Canada, Ltd.

^{*}Chairman

[†]Argonne Coordinator

LIST OF PANELS AND PARTICIPANTS

VII. Magnetism

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R. M. Moon	Oak Ridge National Laboratory
C. G. Shull	Massachusetts Institute of Technology
E. D. Thompson	Case Western Reserve
W. G. Williams	Rutherford Laboratory, U.K.
W. B. Yelon	University of Missouri-Columbia

VIII. Neutron Sources

J. M. Carpenter *	Argonne National Laboratory
A. S. Arrott	Simon Fraser University
M. Barbier	MITRE Corporation
R. G. Fluharty	Los Alamos Scientific Laboratory
M. Kimura	Tohoku University, Japan
E. W. J. Mitchell	University of Reading, U.K.
M. S. Moore	Los Alamos Scientific Laboratory
R. W. Pelle	Oak Ridge National Laboratory
G. J. Russell	Los Alamos Scientific Laboratory
N. Watanabe	Tohoku University, Japan

* Chairman

† Argonne Coordinator

LIST OF PANELS AND PARTICIPANTS

IX. Radiation Effects

P. Sigmund*	University of Copenhagen, Denmark
T. H. Blewitt†	Argonne National Laboratory
R. C. Birtcher	Argonne National Laboratory
B. S. Brown	Argonne National Laboratory
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J. Diehl**	Max Planck Institut, Stuttgart, Germany
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M. A. Kirk	Argonne National Laboratory
C. Y. Li	Cornell University
R. K. Merkle	Argonne National Laboratory
F. Nolfi, Jr.	Argonne National Laboratory
P. J. Persiani	Argonne National Laboratory
M. T. Robinson	Oak Ridge National Laboratory
T. L. Scott	Argonne National Laboratory
J. L. Warren	Los Alamos Scientific Laboratory
M. S. Wechsler**	Iowa State University
H. Wiedersich	Argonne National Laboratory
F. Wiffen	Oak Ridge National Laboratory

* Chairman

** Subgroup Co-chairmen

† Argonne Coordinator

PROGRAM FOR THE CONFERENCE AND WORKSHOP ON USES OF
ADVANCED PULSED NEUTRON SOURCES

Sunday, October 19, 1975

Informal Reception, sponsored by the University of Chicago and AUA

Cortez Room, Ramada Inn

7:00 - 10:00 p.m.

Monday, October 20, 1975

Conference on Uses of Advanced Pulsed Neutron Sources

Argonne National Laboratory

ZGS Auditorium, Building 362

9:00 a.m.	<u>Welcoming address</u>	R. G. Sachs, Director Argonne National Laboratory
	<u>Survey Talks</u>	
9:15 a.m.	Review of Neutron Sources	E. W. J. Mitchell University of Reading, U.K.
10:00 a.m.	Coffee Break	
10:15 a.m.	Solid State Physics	R. J. Birgeneau Massachusetts Institute of Technology
11:00 a.m.	Chemistry	J. W. White Institute Laue-Langevin, France
12:15 p.m.	Lunch	Bldg. 213 - Cafeteria Private Dining Room C
1:30 p.m.	Physics of Liquids	K. Sköld Argonne National Laboratory
2:15 p.m.	Biology	B. P. Schoenborn Brookhaven National Laboratory
3:00 p.m.	Coffee Break	
3:15 p.m.	Computer Simulation	A. Rahman Argonne National Laboratory
4:00 p.m.	Radiation Effects	T. H. Blewitt Argonne National Laboratory
5:30 p.m.	Dinner	Building 213 - Cafeteria

Description of Sources

7:00 p.m.	Potential Use of ORELA with Booster System	R. W. Pelle Oak Ridge National Laboratory
7:45 p.m.	LAMPF Weapons Neutron Research Facility	R. G. Fluharty Los Alamos Scientific Laboratory
8:30 p.m.	ANL Intense Pulsed Neutron Source	J. M. Carpenter Argonne National Laboratory

Tuesday-Friday, October 21-24, 1975

Workshop on Advanced Pulsed Neutron Sources

Argonne National Laboratory

Tuesday, October 21, 1975

Plenary session:

9:00 a.m.	Welcoming address	D. L. Price
9:05 a.m.	Commission to workshop panel members	S. A. Werner

Parallel sessions:

1. Presentation of data on pulsed slow neutron sources J. M. Carpenter
2. Presentation of data on fast neutron irradiation facilities T. H. Blewitt

10:00 a.m. Coffee Break

10:15 a.m. - 5:00 p.m. Individual panel meetings

6:00 p.m. Social hour (cash bar) Building 213 - Cafeteria

7:00 p.m. Banquet, sponsored by the University of Chicago and AUA Building 213 - Cafeteria

Speaker:

T. H. Fields

Associate Laboratory Director for High Energy Physics, ANL
Title: Shared Use of High Energy Physics Facilities

Wednesday, October 22, 1975

9:00 a.m. - Individual panel meetings
12:00 noon
1:00 p.m. Plenary session -- brief reports from panel chairmen
2:00 p.m. - Individual panel meetings
5:00 p.m.

Thursday, October 23, 1975

9:00 a.m. - Individual panel meetings
12:00 noon
1:00 p.m. Plenary session -- discussion on user policy
2:00 p.m. Individual workshop panels to draft reports summarizing
their ideas, findings and conclusions

Friday, October 24, 1975

Plenary session:

9:00 a.m. Reports from panel chairmen
11:30 a.m. Workshop summary S. A. Werner
12:30 p.m. Workshop adjourns

I. REPORT OF THE PANEL ON BIOLOGY AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. INTRODUCTION

We have reviewed existing applications of neutron diffraction in biology and the prospects opened up by IPNS in that field. We wish to specify that we have restricted our attention to molecular biology and recognize that many significant problems may have been overlooked. Application of neutron diffraction to biological problems is quite recent, but it is apparent that the potential of the technique has been barely tapped. See the review by Schoenborn and Nunes.¹

Biological processes involve the interaction of various macromolecular species such as proteins, nucleic acids and lipids with each other, smaller organic molecules, water and salts. Detailed study of the basic processes seeks knowledge of the kinetic pathways by which reactions occur and the structure of the interacting components. Though many techniques are employed, x-ray and neutron scattering provide the most direct and detailed information concerning molecular architecture, and are potentially useful in studies of reaction kinetics. Neutron work in biology has recently enjoyed a great expansion because (as is described below) it provides more information about a system than any other single technique, and involves less perturbation of the system under study than experiments using ionizing radiation (e.g. x-rays, or electron microscopy).

Diffraction or scattering techniques are applicable to studies of a surprisingly large number of biological components or assemblies because of their inherent paracrystalline order. Thus muscle, collagen, myelin and retinal segments are functionally ordered while proteins, nucleic acids, various membranes, viruses, lipids, etc., can be induced to form ordered arrays by appropriate treatment. The presence of order in a sample permits the recording of a diffraction pattern from which the dimensions of a repeat unit in the sample may be determined and structural information may be deduced. The quality of the structural information depends on the degree to which the specimen is ordered. In principle, three-dimensional order will permit determination of the atomic arrangement, paracrystalline order provides molecular group information, while small angle scattering from solutions of macromolecules makes possible determination of particle size and shape. When differential labeling of components is possible small angle scattering can yield the spatial distribution of the subunits of a macromolecular assembly.

B. NEUTRON AND X-RAY COMPARISONS

There are at present two general domains in which diffraction techniques are significant in molecular biology: Single crystal structure determination of macromolecules and low angle diffraction of solutions and ordered but non-crystalline systems. Neutrons offer a number of advantages over x-rays in both areas.

For protein structure studies the significant neutron properties are: (1) the scattering length of an atom for a neutron depends chiefly upon the neutron-nucleus interaction, and covers a narrower range than that of x-rays, (2) chemically similar isotopes may have very different neutron scattering lengths, permitting tagging or exchange studies which do not otherwise affect delicate protein machinery, and (3) thermal neutrons induce little or no damage when they interact with biological materials. Table I compares neutron and x-ray scattering lengths for the atoms commonly found in organic

Table I. Neutron and x-ray scattering factors for elements and isotopes commonly present in biological molecules.

Element	Atomic No.	Neutrons $b(10^{-12}\text{cm})$	X-rays $f_x(10^{-12}\text{cm})$	
			$\sin\theta/\lambda = 0$	$\sin\theta/\lambda = 0.5 \text{ \AA}^{-1}$
H	1	-0.374	0.28	0.02
D	1	0.667	0.28	0.02
C	6	0.665	1.69	0.48
N	7	0.940	1.97	0.53
O	8	0.575	2.25	0.62
Na	11	0.351	3.09	1.14
P	15	0.51	4.23	1.83
S	16	0.285	4.48	1.9
K	19	0.35	5.3	2.2
Ca	20	0.49	5.6	2.4

material. The neutron scattering length of hydrogen atoms is of the same magnitude as the other atoms; therefore, neutrons can detect and localize these atoms which, through hydrogen bonding structures, play a very important role in the stabilization of protein structure and protein function. Furthermore, the hydrogen scattering length is negative in phase while that of deuterium and most other atoms is positive. This heightens the contrast of a neutron Fourier map compared with that of x-rays, and can simplify interpretation of a low resolution map. If deuterium is partially substituted for hydrogen there is a maximum contrast between the regions in which exchange has or has not occurred which is of additional value in determining accessibility of protein components to the solution. An additional very significant datum for large molecule studies is the nitrogen scattering length ($0.94 \times 10^{-12} \text{ cm}$) which makes it easily distinguishable from carbon or oxygen in a neutron fourier map allowing identification and orientation of certain amino acid side chains (e.g. histidine, asparagine, glutamine, etc.) with a speed and certainty impossible with x-ray data.

For small angle scattering studies where identification of molecular groups is the main object, one is interested in the density contrast which can be introduced into these groups and utilized for identification. The appropriate x-ray and neutron property to be used for comparison is the scattering amplitude density. This density is equal to the product of the scattering length (or amplitude) per atom times the number of atoms per unit volume summed over all atoms in the molecular group.

The value of ϕ_x , the scattering amplitude density for x-rays is always positive and ranges, for biomolecular components, from $0.7 - 1.5 \times 10^{11} \text{ cm}^{-2}$ (see Ref. 2). For neutrons where hydrogen scatters with negative amplitude, ϕ_n can take negative as well as positive values. When deuterium, which scatters with positive amplitude, is substituted for hydrogen, ϕ_n increases thus introducing a density contrast with the hydrogenous material. Large

density contrasts can be introduced easily by replacing hydrogen which is not bonded to carbon with deuterium when placed in heavy water. Table II (see Appendix) lists the scattering densities of a number of biological substances. Neutron scattering densities of these substances can be controllably varied by substituting deuterium for hydrogen. If deuterated material is used, it is possible to vary the scattering density to one component of a molecule or by simply varying the H_2O/D_2O ratio of the solvent. For example, the protein or nucleic acid of a virus will effectively disappear when deuterated and then be determined by the other unmatched components. Alternatively, deuterating one component of a complex will be a design of experiments. These very powerful techniques are being exploited in studies of myelin, viruses, collagen, muscle, and others (see *Proc. Natl. Acad. Sci. Biol.* 27, in press).

Though one can vary the x-ray scattering density of appropriate solutes, one cannot cover a wide range of biologically interesting substances. Large variations in scattering density of a macromolecule is achieved by chemical modification.

Table II. X-ray and neutron scattering at low angles for biologically important substances

Molecular Component	S
	Neutron
Water H_2O	-0.55
Heavy Water D_2O	+6.34
Protein (H)	+3.44
Protein (D)	+8.24
Nucleic Acid (H)	+4.14
Nucleic Acid (D)	+7.34
Fatty Acid (H)	-0.55
Fatty Acid (D)	+6.34
Carbohydrate (H)	+4.14
Carbohydrate (D)	+8.24

C. CURRENT APPLICATIONS

1. Neutron Diffraction and Protein Structure

The feasibility of neutron diffraction studies of protein crystals has been established, and the results are very encouraging.

Three-dimensional neutron crystallographic data have been collected to 2.0 and 1.8 Å resolution on met-myoglobin and CO myoglobin respectively at Brookhaven National Laboratory (Ref. 4). While structure refinement is still under way, the maps (phased from the x-ray determined structure) do reveal hydrogen atoms, deuterium replaced sites, and surface water, as well as structural details visible in the x-ray maps. The neutron maps also seem much sharper than the x-ray maps of comparable resolution. Surface side chains, often exhibiting very large disorder in the x-ray maps, are often quite clear in the neutron maps. This is likely due to the absence of radiation damage by neutrons, a fact which may be very valuable in studies of radiation sensitive crystals; e.g., t-RNA and actin. In the Brookhaven studies, no sample deterioration was detected after six years (approximately six months of actual exposure in a 10^7 n/cm² sec flux).

The chief difficulties in neutron studies of protein single crystals are the fluxes available at existing reactor facilities, the difficulty of obtaining large crystals, and presently available instrumentation. Each of the two studies cited above required crystals of approximately 27 mm³ in volume and roughly three months data collection time at one of the world's most intense neutron sources. The instrument used in this work was a single detector automatic diffractometer. Six to seven thousand independent reflections were measured. It has been estimated that use of a position sensitive detector, or array of detectors, may increase data collection rates by factors of 10 or 20.

Following are the types of studies that can be performed with neutron diffraction of protein crystals which will add to our knowledge derived by x-ray diffraction.

1. Determination of the H atom positions: this will make it possible to describe the detailed hydrogen bonding within the protein. The knowledge of the H positions will better characterize amino acid residues with structural or functional importance. It will also be possible to follow structural changes as a function of pH.

2. Determination of water structure: protein crystals contain over 40% solvent, concerning which little structural information is available. One wishes to know the water molecule arrangements both in the ordered region near the protein surface and also at greater distances where less order is expected.

3. A more accurate determination of the molecular structure, which will be useful in theoretical calculations involving proteins. Since there is little radiation damage and all the data can be collected on one crystal, the quality of the diffraction data and resulting structure should be high.

4. Study of the binding of small molecules in the crystals: this is of special interest in enzyme and antibody studies. Because there is no radiation damage the same crystal can be used for both native protein and for derivative studies thus eliminating scaling errors. H-D substitution may be exploited to "tag" small molecules, making them more visible in difference maps.

2. Neutron Anomalous Scattering

The solution of protein structure by x-ray methods depends largely on the heavy atom isomorphous replacement technique for phase determination. To a lesser extent anomalous dispersion effects are used to aid in the phase determining process. In the neutron case a few nuclei (e.g., Cd¹¹³, Sm¹⁴⁹, Gd¹⁵⁷) have very strong resonances in the thermal region and display anomalous effects larger by an order of magnitude than those available in the x-ray case. These effects include wavelength sensitive scattering amplitudes which are both unusually large and strongly phase shifted. As a result these nuclei serve both as heavy atoms and as anomalous scatterers and in principle, should be substantially more powerful for phase determination than available x-ray methods. Full utilization of the promise of this technique requires high neutron fluxes at relatively short wavelengths ($\sim 1 \text{ \AA}$). Since the IPNS pulsed source is particularly rich in epithermal neutrons and provides simultaneous measurement at many wavelengths, it would appear to be well suited for exploitation of the potential of neutron anomalous scattering techniques in large molecule structure determination.

3. Neutron Small Angle Scattering Studies

The application of neutron small angle scattering to biological problems is of very recent origin and is based largely on the great difference in scattering lengths for H and D, a difference that doesn't exist in the x-ray case. Deuterium substitution makes possible both large contrast variation to assist in group identification and also differential labeling of subgroups in large molecular aggregates such as ribosomes.

The first small angle scattering studies of the nerve myelin sheath by neutrons were carried out as recently as 1969.⁵ More recent studies⁶ have yielded much precise information concerning the structure and water distribution of the sheath. The myelin studies were carried out on freshly dissected nerve which is a highly ordered material. The sheath consists of a spiral wrapping of a membrane pair which surrounds the central nerve axon and serves as an insulating barrier. The neutron results demonstrate that the space between the membrane bilayers which is water rich is comparable in thickness to the water-excluding membrane material. Additional experiments testing the effects of reagents such as DMSO, which block nerve conduction, suggest in concurrence with x-ray findings that a reversible structural transformation occurs and that it is mainly associated with water removal from the spaces between membrane layers. Correlated x-ray and neutron data were used very successfully in these studies.

It is very significant that neutron small angle studies on macromolecules in solution also provide valuable microscopic data. Human serum lipoproteins are water soluble particles in which serum lipids are associated with protein components. The lipid fractions contain long hydrocarbon chains which are

nonpolar and have low scattering density for neutrons. The protein components which have a higher proportion of polar groups and also a higher scattering density can thus be distinguished from the lipids by their scattering properties. Lipoproteins are involved in a variety of biological processes including the maintenance of cell membranes, lipid transport and energy metabolism. Knowledge of particle size and shape and the distribution of lipid and protein components can lead to greater understanding of how these molecules function. In a recent neutron scattering study⁷ of a series of solutions of a lipoprotein in various H₂O-D₂O mixtures much of this information was obtained. The basic findings were that the particles were quasi-spherical, that the particle volume was $\sim 3.10^6 \text{ \AA}^3$ and that the volume fraction occupied by hydrocarbon was 60%. Also the centers of gravity of the hydrocarbon and the polar regions coincided but the radius of gyration of the polar region was larger. Thus the core of the particle is predominantly occupied by hydrocarbon chains, while the protein components are concentrated in the outer shell regions. Thus a rather detailed picture of the structure and organization of a human serum lipoproteins emerges. Although x-ray small scattering can, in principle, provide some of the same information it is obtainable only with great difficulty and the relative location of hydrophilic and hydrophobic regions would be ill defined at best. The neutron studies can be carried out more quickly and precisely and they provide better discrimination.

A third type of neutron small angle scattering study, involving differential labeling,^{8,9} is attracting strong attention because of its potential for 3-dimensional mapping of the distribution of the fifty or so individual protein molecules that make up the two subunits of the ribosome. Protein synthesis takes place at the ribosome and mechanistic studies of the process are of current high interest. The isotopic labeling technique involves the incorporation of a pair of deuterium rich proteins into the otherwise normal ribosome subunit and the measurements of the neutron small angle scattering from a solution of ribosome subunits. The neutron scattering curve then gives the distance between the centers of mass of the two labeled proteins. If this is repeated many times the distances between all possible pairs can be determined and a triangulation procedure leads to the complete mapping. Such studies are currently under way at both Brookhaven and Grenoble.¹⁰

These are just a few examples of the application of a rapidly developing technique which has great potential for biology. The main limitation at present is the availability of appropriate instrumentation for the measurements. Essentially all of the biological experiments are being carried out at three sites, Brookhaven, Harwell, and ILL - Grenoble, and only at the latter site is there a highly developed instrument. As a result only a small fraction of the current demand for neutron data on biological materials is being met. (For a description of a small angle scattering instrument for a pulsed source see the report of the panel on the Structure of Disordered Solids.)

4. Availability of Highly Deuterated Samples

One of the key requirements for much of the neutron small angle scattering studies is the availability of highly deuterated samples or sample components. While partially deuterated samples can often be prepared easily by D₂O exchange, fully deuterated biological samples are prepared only with some difficulty. One of the prime sources of such material is the algae

farm¹¹ at Argonne where plant microorganisms are grown in D₂O and fully deuterated proteins and other bio-macromolecules are extracted from the algae, yeast or other microorganism. The Argonne laboratory is involved in collaborative experiments with groups at the Max Planck Institutes in Munich and Berlin and ILL - Grenoble on the application of the label triangulation method to the 50S subunit of E. Coli ribosomes. Measurements of several inter-protein distances are currently under way.¹² In another effort the structure of chlorophyll oligomers is being investigated by small angle neutron scattering at Harwell in a joint effort with the Argonne group. These oligomers which form spontaneously in nonpolar media and reach molecular weights > 20,000 are believed to serve as good models for antenna chlorophyll in green plants. Here again the availability of fully deuterated samples is expected to be the key to the success of the project.

D. LONG RANGE POTENTIAL

The Intense Pulsed Neutron Source could open up new frontiers for neutron diffraction studies of the kinetics of biological structural changes as well as enabling extension of elastic scattering studies in directions not now currently feasible. The utility of this facility can be most concretely evaluated in relation to the biological structural problems currently under investigation at the Brookhaven National Laboratory and the Institute Laue-Langevin. The biological structures can be considered in three broad categories: protein and nucleic acid crystals; periodic cellular assemblies; and solutions of proteins and complex aggregates.

1. Protein Crystallography

At present, the crystal structures of two forms of myoglobin are being refined by neutron crystallography, and neutron diffraction data are being collected from triclinic lysozyme. This compares with about 50 protein crystal structures solved to high resolution by x-ray diffraction analysis and another 100 or so under study. The most distinctive features of the neutron scattering density maps are the clearly visible protons which can hardly be seen by x-rays and the distinguishability of the protons which can be exchanged with deuterons. Although it would not be feasible to consider studying with neutrons every protein analyzed with x-rays, it is evident from the present neutron diffraction results that more proteins should be studied and that new information can be derived. Such an expanded effort would require the increase in the speed of data collection possible with the time-of-flight spectrometer planned for the IPNS compared to that with the diffractometers currently in use at BNL and ILL.

The biological structure problems where neutron crystallography could provide novel information include proteins which can be cocrystallized with small ligands and complex assemblies. Antigen-antibody and model substrate enzyme complexes have been crystallized. In systems like these the two components can be differentially labeled with H or D. Static elastic scattering studies would clearly reveal the structural interactions. In some cases the kinetics of binding or structural change could be followed in the crystalline state. With protonated ligand-deuterated protein, it would be possible to characterize the dynamics of the interaction by inelastic scattering measurements.

2. Macromolecular Complexes

Neutron crystallography of macromolecular assemblies such as viruses and enzyme complexes has not yet started for lack of suitable sources and instruments. A high intensity pulsed source with long wavelength neutrons would make studies on crystals of such large structures possible. A distinctive advantage of neutrons over x-rays in such studies would be the ability to distinguish components with large scattering density differences such as nucleic acid and protein in low resolution density maps. The combination of neutron and x-ray measurements on virus crystals, for example, even at relatively coarse spatial resolution should reveal significant details of the protein nucleic acid interaction.

3. Periodic Cellular Assemblies

The high degree of molecular order in cellular structures such as muscle, tendon, nerve myelin sheath and retinal rod photoreceptors makes it possible to carry out diffraction studies on intact cellular structures. Interpretation of diffraction patterns from such complex structures requires coordinated electron microscopic and chemical studies. The combination of neutron and x-ray diffraction data from membrane structures is providing a new and powerful method of ultrastructural analysis. By contrast variation the distribution of water, protein and lipid is being mapped. The disorder and peripheral structures in such cellular assemblies present challenging problems both in specimen preparation and in the design of diffraction apparatus. It is clear from past experience that the neutron diffraction equipment developed for study of large period structures can be applied to study of the molecular organization in living tissue. The general instrumental needs are high intensity and high spatial resolution, but each biological system presents special problems which require flexibility in the instrumentation.

4. Dynamics of Living Processes

A prime objective of structural studies on intact tissue is the characterization of the dynamics of living processes. Kinetic studies of muscle contraction are in progress with synchrotron radiation. With a pulsed neutron source complementary neutron diffraction could be undertaken. Since this is a developing field of study it is difficult to project what will be the most critical questions for investigation with neutrons when a suitable pulsed source is available. An important limitation to many kinetic experiments is the difficulty of providing synchronization of the reaction of interest. If this obstacle can be overcome, there are many intriguing areas of investigation worthy of consideration such as the release of DNA from λ phage. Neutron scattering studies on solutions of proteins and macromolecular assemblies bring together the methods of biochemistry, macromolecular physical chemistry and cell biology. The contrast matching technique for solutions in different D_2O-H_2O mixtures has been developed to the point that small angle neutron studies on solutions are providing more information more rapidly than can be obtained by x-ray scattering. An important area for development in small angle scattering will be the study of the kinetics of reactions in solution. Since substrates and other ligand molecules can be differentially labeled, it should be possible to follow the rate of structurally important changes in interactions with enzymes or regulatory proteins.

5. Mapping of Organelles

The neutron scattering studies on solutions of differentially labeled ribosomes is a dramatic illustration of the kind of information obtainable with neutrons for which there is no comparable x-ray equipment. With improvements in instruments and methods of differential labeling such topological studies will be extended to a great variety of complex biological structures.

6. Mechanisms of Regulation

The most critical problems in structural biology relate to mechanisms of regulation; for example: the release of Ca from the sarcoplasmic reticulum following nerve excitation; the transmission of the nerve impulse by release of neurotransmitters at the presynaptic membrane; the depolarization of the postsynaptic membrane by binding of neurotransmitters to a specific receptor and the control of muscle contraction by the binding of Ca by regulatory proteins in the thin filaments. There are general similarities in very different biological control processes such as vision, hormone action and gene regulation.

The active proteins have been isolated from many of these systems. In the case of membrane structures, proteins such as rhodopsin from retina, the acetylcholine receptor of the postsynaptic membrane, the Ca dependent ATPase of the sarcoplasmic reticulum, the cytochrome oxidase and ATPase of mitochondria, etc., have been isolated and reconstituted with lipids in active forms. Neutron diffraction studies have been started on some of these systems. The high intrinsic scattering density contrast of the component parts, as well as the feasibility of H-D labeling, make it possible to carry out structural and kinetic studies by neutron diffraction which have no direct parallel with x-ray studies. Similar incisive studies can be carried out with nucleoproteins such as viruses, chromatin and ribosomes.

With suitable sources, neutron diffraction will become a technique of importance in structural biology comparable to that of x-ray diffraction, NMR spectroscopy, and electron microscopy. The major advances in the application of neutron diffraction to biology will undoubtedly resemble the advances in the application of other physical techniques: progress follows the development of the technology and the control of the biological material, but the breakthroughs are the result of new insights which make unanticipated use of the systematic development of tools and methods.

7. Some Specific Problems

We will mention a few specific problems that would benefit from the high neutron fluxes available from a pulsed source such as IPNS.

1. The mechanisms of action of enzymes such as serine proteases would be further clarified by high resolution neutron studies which could locate labile protons in the active site.

2. Electron transport proteins such as the cytochromes are relatively small proteins of high interest which could be easily handled at IPNS.

3. Low resolution studies of the structure of t-RNA would be useful in conjunction with sequencing and x-ray structure work in progress. It is of great interest to see if the limitation of 3 Å resolution in the x-ray work is due in part to radiation damage and can be overcome in a higher resolution neutron study.

4. Two types of human serum protein, a high and a low density lipoprotein, are available in large quantity from the University of Chicago. Studies of the intact complexes, of enzyme modified products, of reassembled materials and of the effects of activators and inhibitors on the interaction of lipolytic enzyme with the lipoproteins are all of high interest.

The above-mentioned research problems may well be addressed at currently available neutron sources. Some more speculative experiments which will probably await higher intensity sources include the following.

1. Titration of all groups at the protein surface, by solution of the three dimensional structures of the protein at various pH values.

2. Determination of spin density around magnetic atoms in iron-containing heme and non-heme proteins.

3. Application of inelastic scattering to a deuterated protein with only one amino acid or ligand hydrogenated in the active site; the inelastic spectrum would be dominated by the motion of this group, and the data would be relatively simple to interpret.

4. Study of the kinetics of the folding of a protein through H-D exchange. Chemical reduction resulting in denaturation and subsequent reoxidation has in one case (ribonuclease) been demonstrated to result in a high degree of renaturation (as measured by enzyme activity) within three hours. Other proteins can be unfolded and refolded through application of similar techniques.

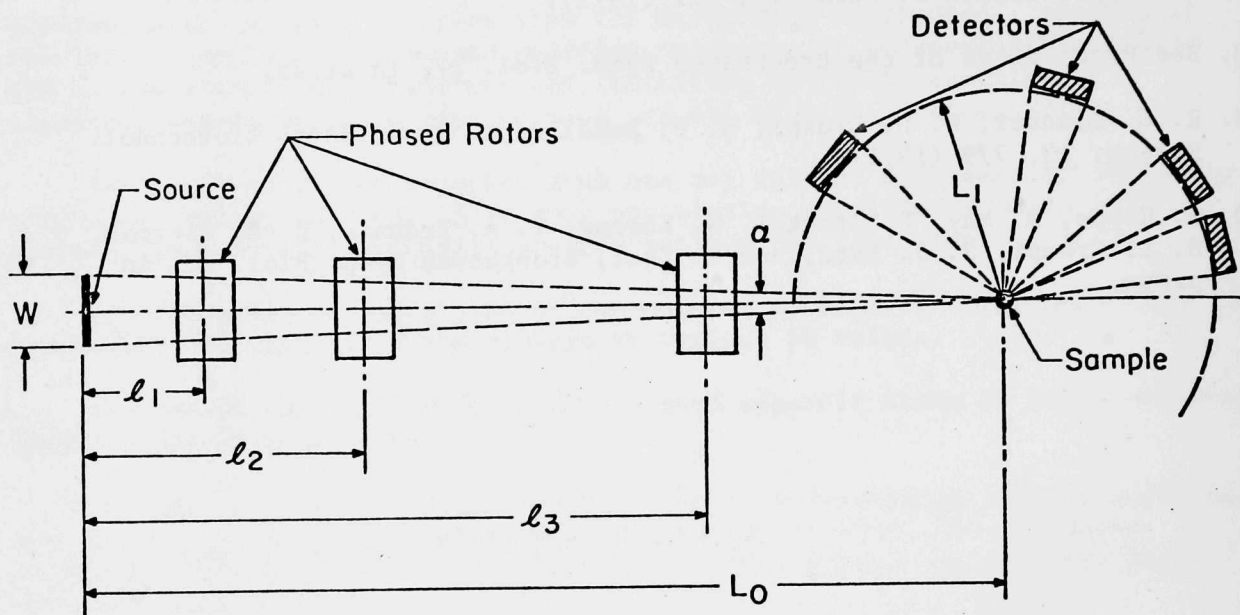
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Appendix I. Description and Performance Estimates of a Possible TOF Single Crystal Diffractometer for IPNS.

Fig. 1. TOF-Single Crystal Diffractometer



The classical white beam Laue technique appears to be ideally adapted to exploit the full potential of pulsed neutron beams in single crystal diffraction. In this method, a pulsed polychromatic neutron beam impinges on the single crystal sample, simultaneous diffraction from many reciprocal lattice points occurs and the diffracted neutrons are detected with a large area position sensitive detector using time-of-flight analysis. Thus each r.l.p. will be characterized by x , y and t (time) coordinates which will suffice to both identify the direction of the diffracted beam and resolve the orders. The crystal remains stationary while the intensities are measured since the wavelength distribution provides the necessary integration over the crystal mosaic spread. Data collection at a small number of fixed orientations should provide the entire 3-dimensional set of Bragg intensities.

The basic layout of the instrument is shown in Fig. 1. Three phased choppers are provided; the one nearest the source is to reduce the high energy direct beam background and the next two to limit the wavelength range in order to prevent frame overlap. The path lengths are fixed by the burst width of the neutron pulse and by the resolution required in order to resolve the most closely spaced reciprocal lattice points for the most difficult class of problems to be attempted. The equations which relate path length to the time-of-flight uncertainty and to wavelength are:

$$L_0 + L_1 = a/(\Delta t/t)$$

where L_0 is source to sample distance, L_1 is sample to detector distance, $\Delta t/t$ is the fractional uncertainty in neutron travel time due to the neutron burst width Δt and a is a constant relating burst width to neutron velocity, $a = v\Delta t$ and $0.03 \lesssim a \lesssim 0.11$ m. In calculations below, we adopt $a = 0.11$ m.

The range of neutron wavelengths which can be utilized with no frame overlap is

$$\lambda_{\max} - \lambda_{\min} = \frac{3960 \text{ Å-m/sec}}{f(L_0 + L_1)}$$

where f is the repetition rate. The chopper transmission must be phased to pass this wavelength range.

The resolution in Q depends on w/L_0 and w'/L_1 , the angular divergences due to source width and detector element width as well as $\Delta t/t$, the time uncertainty due to burst width. At $\theta = 90^\circ$ (where resolution requirements are most severe)

$$\Delta Q_x = \Delta Q_y = (w/L_0 + w'/L_1) |\vec{k}| \text{ and } \Delta Q_z = 2k \frac{\Delta t}{t}$$

(where \vec{k} is the wavevector). The basic resolution criterion is that the maximum resolution element extension is smaller than $2\pi\tau_{\min}$ ($\tau = 1/d_{hk}$). If we arrange that the several resolution elements make equal contributions, this leads to

$$\Delta t/t = 1/\sqrt{3} \cdot \lambda_{\min}/\lambda_{\max}$$

for the back scattering direction where resolution requirements are most severe.

We now discriminate amongst three classes of crystals which might be investigated. They differ in maximum unit cell dimension (assumed cubic) and the order n to which reflections are to be measured. Thus, they correspond roughly to large biological, intermediate organic and smaller inorganic unit cells. The table lists some pertinent parameters for these cases.

Table I

Class	d_{100}	n	$2\pi\tau_{\min}$	τ_{\min}/τ_{\max}	λ_{\min}	λ_{\max}
1	100 Å	50	0.063 Å ⁻¹	0.020	4.04 Å	10.0 Å
2	25	30	0.25	0.033	1.66 Å	8.56 Å
3	10	20	0.63	0.050	1.00 Å	7.90 Å

Choosing $\tau_{\min}/\tau_{\max} = 0.02$ for class 1 to define the resolution required leads to

$$\Delta t/t = 0.0115 ,$$

$$L_0 + L_1 = 9.6 \text{ m} \quad \text{and}$$

$$L_0 \approx 9 \text{ m} \quad \text{and} \quad L_1 \approx 0.6 \text{ m} .$$

This also fixes the source width w at 10 cm and the detector element width w' at 0.6 cm. Thus rather modest flight paths will suffice to resolve neighboring reflections in quite complex crystals. Also the sample-detector distance is kept reasonably small which in turn keeps the detector from becoming too large. Note that no allowance is made for sample mosaic spread or sample size in the resolution requirements. The wavelength range available becomes fairly restrictive for class 1 measurements, (λ_{\min} is fixed by requiring that d_{\min} be measured at $\theta = 90^\circ$) and to collect all data a number of crystal orientations will be required. Shifting the wavelength range to give better coverage will increase the resolution requirements so little if anything is gained.

The number of reflections simultaneously excited by a beam of a given wavelength spread can be easily estimated using the Ewald Sphere construction in reciprocal space. Figure 2 depicts this construction for both a slice in reciprocal space due to a normal monochromatic beam ($\Delta\lambda = 0.02$) and for a broad spectrum beam. All reflections within the shaded areas of the two constructions will be simultaneously excited. The advantage the TOF over the monochromatic technique lies largely in the magnitude of this ratio of simultaneously excited reflections. Estimates of the number of simultaneous reflections which will be detected with a $21 \times 21 \text{ cm}^2$ position sensitive detector (900 detector elements) is given in Table II for the three classes of problems mentioned. The table also lists, for the λ -range which allows measuring d_{\min} for the class in back scattering, Q_{\max} , d_{\min} and d_{\max} .

The measuring time T will be determined by the accuracy required for reflections near $Q = 2\pi\tau_{\max}$ where the structure factors and signal to background ratio S/B are small. We shall assume that $S/B \approx 1/20$ for such reflections and that the precision required is $\sim 30\%$. Then estimating the signal rate and background count rate for the proposed instrument, we can calculate data collection time.

The count rate for a coherent elastic reflection turns out to be

$$\frac{\phi(\lambda_0) \lambda^4 N_s \left(\frac{d\sigma}{d\Omega}\right) k_0}{V_c \cdot 2\sin^2\theta}$$

where $\phi(\lambda_0)$ is the flux per unit of wavelength, N_s is the number of nuclei in the specimen, $d\sigma/d\Omega$ is the coherent elastic cross section, k_0 is the incident wavevector, θ the Bragg angle and V_c the unit cell volume.

The background count rate observed in a resolution element Δ^3Q_{res} is

Table II

Class 1

θ	Q_{\max}	d_{\min}	d_{\max}	N
15°	0.81 \AA^{-1}	8	22	23
30°	1.57	4	11	318
45°	2.22	2.9	8	1270
60°	2.72	2.4	6.6	2870
90°	3.14	2	5.5	5100

Class 2

15°	1.95	3.3	16.8	5
30°	3.77	1.65	8.4	69
45°	5.33	1.2	6.1	275
60°	6.53	1.0	5.1	620
90°	7.54	0.83	4.2	1102

Class 3

15°	3.25	1.93	15.2	1.5
30°	6.28	1.00	7.9	20
45°	8.89	0.70	5.5	82
60°	10.88	0.58	4.5	106
90°	12.57	0.50	3.9	327

$$B = C^{\text{Incoh}} + C^{\text{coh, inel}} = \phi(\lambda_o) \frac{\lambda_s^4 N_s}{(2\pi)^3} \frac{\Delta^3 Q}{2 \sin^2 \theta} \left(\frac{\partial \sigma^{\text{Inc}}}{\partial \Omega} + \frac{\partial \sigma^{\text{coh, inel}}}{\partial \Omega} \right)$$

The flux at the sample per unit wavelength is

$$\phi(\lambda) = \frac{f}{L_1} I(\lambda)$$

where $f = 60$ Hz, $L_1 = 9$ meters and $I(\lambda)$ is the number of neutrons per steradian per unit wavelength per pulse. The latter quantity has been evaluated for a $2'' \times 10$ cm \times 10 cm Be reflected moderator at IPNS.

In a measurement the signal rate S and the background rate B are measured. S is obtained from signal plus background in one resolution element and background may be obtained from n neighboring elements. Then in a counting time T , the statistical error in S is

$$\frac{\partial S}{S} = \frac{1}{S/B} \frac{1}{(BT)^{1/2}} \cdot (1 + S/B + 1/n)^{1/2}$$

We have chosen $S/B = 0.05$ and n may be as large as 10. The time to achieve 30% precision will be

$$\frac{\partial S}{S} = 0.30 = \frac{\sqrt{1.15}}{0.05} \cdot \frac{1}{(BT)^{1/2}}, \text{ hence } BT = 5000$$

Thus, the counting time is determined by B . We estimate B assuming $\frac{1}{4\pi} \sigma_{\text{scatt}}$ gives the sum of incoherent and coherent inelastic cross sections. Taking $\sigma_{\text{scat}} = 5b$ and $N = n'V_s$, where $n' \approx 7 \cdot 10^{22}$ atoms/cm³ and V_s is the specimen volume, then

$$B = \frac{\phi(\lambda)\lambda^4\Delta^3Q(7 \cdot 10^{22})(5 \cdot 10^{-24})V_s}{2\sin^2\theta(2\pi)^34\pi}$$

For case 1:

$$Q_{\text{max}} = 3.14, \lambda_{\text{min}} = 4 \text{ \AA}, \phi(\lambda_{\text{min}}) = 2.6 \cdot 10^6 \text{ n/cm}^2\text{sec \AA}$$

$$\Delta^3Q = 4.7 \cdot 10^{-5}, \theta = 90^\circ, \sin^2\theta = 1$$

$$\text{giving } B/V_s = 1.7 \text{ n/cm}^3\text{sec}$$

$$\text{for } V_s = 0.01 \text{ cm}^3, T = 2.8 \cdot 10^5 \text{ sec or } \sim 3 \text{ days}$$

$$\text{for } V_s = 0.03 \text{ cm}^3, T = 10^5 \text{ sec or } \sim 1 \text{ day.}$$

Thus, even for large unit cells data collection times are quite reasonable. Note this is for one sample setting which may produce up to several thousand reflections. Perhaps 5-10 settings may be required. If the sample contains much light hydrogen the above estimate will be quite unsatisfactory. For smaller unit cells, e.g., 10 Å, the time required drops to under an hour; hence, it is clear that such an instrument could gather data very quickly.

The real problems with such an instrument are expected to lie in the data handling and storage. If 10 multidetectors, to efficiently cover all scattering directions, with 100×100 elements each and with 100 time channels were provided, 10^7 memory locations with $10^4 - 10^5$ capacity would be needed for on-line storage. Clearly, a great deal of thought would have to go into planning in order to keep costs within reasonable bounds.

II. REPORT OF THE PANEL ON CHEMICAL SPECTROSCOPY AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. GENERAL INTRODUCTION

Consideration of the projected IPNS II source intensities, particularly in the high energy regime, strongly suggests that it will not only allow considerable expansion of the field of chemical and molecular spectroscopy but will open areas of study which have been previously inaccessible. Figure 1 shows the regions of (Q, ω) space in which excitations occur which are important in the various fields of neutron chemical spectroscopy.

The low energy region should not be dismissed either since it may be that the pulsed operation gives us a new principle for enhancing signal to background in that energy domain. In brief, most of the fast neutrons and γ rays will have flown past the spectrometer by the time the lower energy neutrons of interest are arriving. Much in demand among us is the capability for use of cold neutrons; we would ask that a moderator be provided which is optimized for production of pulses of cold neutrons.

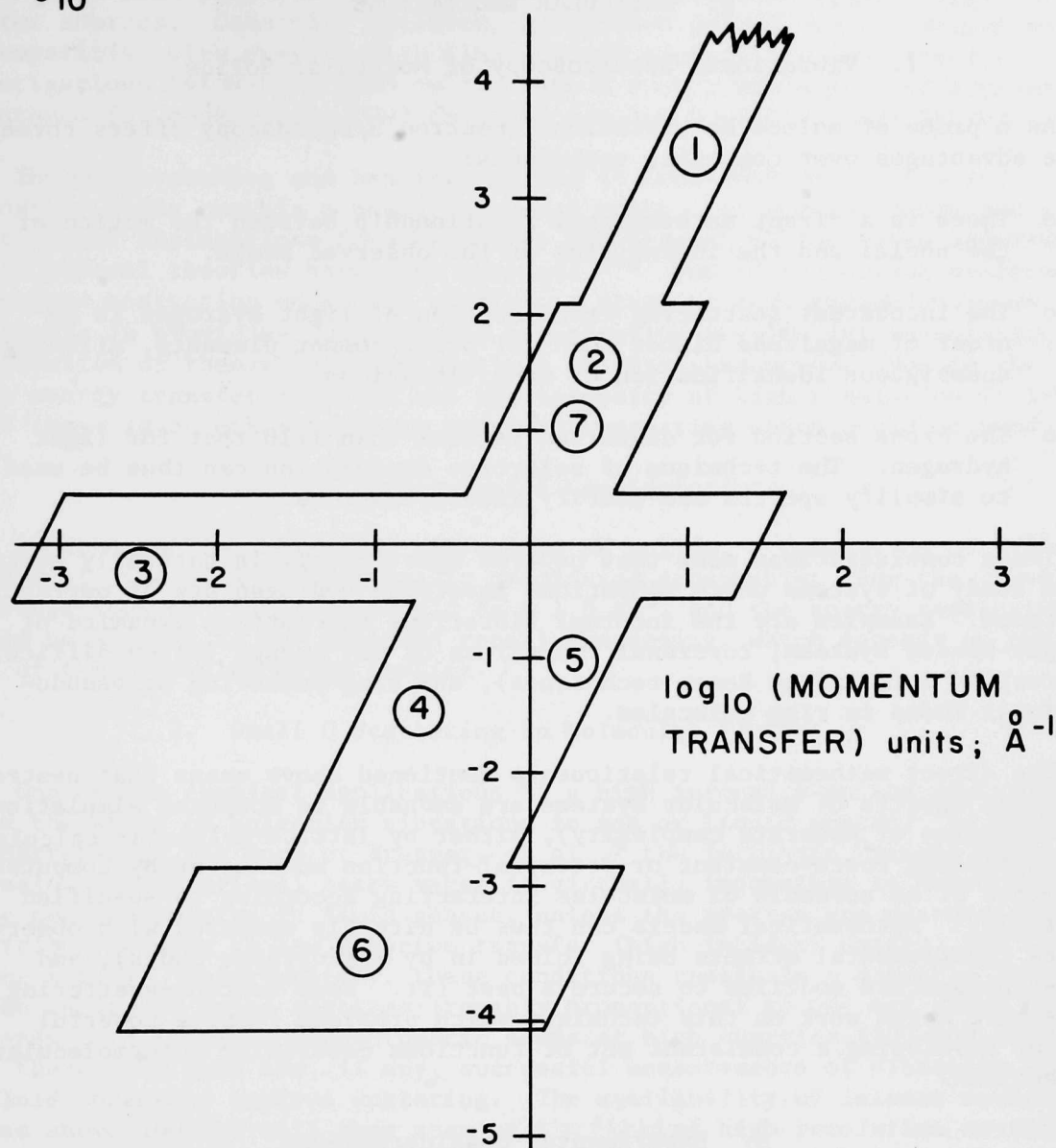
Neutron inelastic scattering techniques are well established for the study of vibrational and rotational motions in hydrogenous molecular systems. This is primarily because of the large incoherent scattering cross section for hydrogen (which varies with incident neutron energy) and because measured neutron spectra provide a direct indication of hydrogen atom motion in these systems. In addition to providing entirely new information, the neutron spectroscopic results are complementary to existing infrared and Raman techniques particularly for cases of optically weak or inactive modes in all types of hydrogen containing systems.

One of the most outstanding virtues of the proposed IPNS II (pulsed) source is that it is possible to capitalize on existing time-of-flight neutron technique and know-how. The higher energy incident neutrons which are available from this source means that it is possible to greatly extend the available range of energy transfers that can be investigated. A number of new fields available through this extension are treated below. Another important development which might prosper with this new source is polarization analysis.¹

For inelastic measurements this technique needs the highest fluxes and the availability of white beam filter polarizers^{2,3} may allow exciting developments to be made. For chemical spectroscopy these are urgently needed; e.g., in surface studies to separate coherent and incoherent scattering. They would also allow construction of very efficient polarized diffractometers and of ultra-high intensity spin echo machines.⁴

Little has been said about such possibilities in the attached report which concentrates mainly on the present and future science that we would like to do. We would strongly support such developments, however.

\log_{10} (ENERGY TRANSFER); units cm^{-1} ($1\text{meV} = 8\text{ cm}^{-1}$)



- ① Intermolecular energy levels
- ② Intermolecular forces
- ③ Low angle "elastic" scattering
- ④ ⑤ Surfaces, adsorbed species, intercalates
- ⑥ Chemical reactions
- ⑦ Biophysical chemistry

Ranges of Energy Transfer and Momentum Transfer of Interest in Various Fields of Chemistry

B. MOLECULAR EXCITATIONS

1. Vibrational Spectroscopy of Molecular Solids

As a probe of molecular vibrations, neutron spectroscopy offers three unique advantages over competing techniques:

- o There is a direct mathematical relationship between the motion of the nuclei and the intensities of the observed peaks.
- o The incoherent scattering cross section of light hydrogen is an order of magnitude higher than for other common elements, allowing unambiguous identification of many vibrations.
- o The cross section for deuterium is less than 1/10 that for light hydrogen. The technique of selective deuteration can thus be used to simplify spectra and clarify interpretations.

These considerations mean that neutron spectroscopy is naturally suited to the study of systems where vibrations involving hydrogen are of particular importance. Examples are the internal vibrations and lattice dynamics of hydrogen-bonded systems, torsional vibrations of CH₃ groups (often difficult to detect by infrared or Raman techniques), and ring-puckering or pseudo-rotational modes in ring molecules.

The direct mathematical relationship mentioned above means that neutron scattering spectra of molecular systems are amenable to computer simulation (for molecules of moderate complexity), either by lattice-molecular calculations utilizing force-constant or potential-function models, or by computer treatments of an ensemble of molecules interacting according to specified potentials.⁵ Mathematical models can thus be directly compared with observed spectra (instrumental effects being folded in by appropriate codes), and assumed parameters modified to secure a best fit. Some neutron-scattering groups have begun work on this technique which promises to be a powerful tool for developing a consistent set of functions describing intermolecular interactions.

a. Experimental Considerations

The large mass of the neutron (compared with that of the photon) means that significant momentum is exchanged in a collision between a neutron and a molecule. In fluid phases this results in broadening of the peaks due to recoil effects. This can be reduced by going to lower scattering angles (see following section for the potential uses of small Q scattering in the spectroscopy of "free molecules") but in general, it is easier to study solids, where the molecule is fixed in the lattice and recoil effects are reduced. With requirements on Q relaxed, a larger range of energy transfer can be utilized at a given count rate.

The addition of a major facility for neutron scattering experiments can be expected to have a beneficial effect in encouraging application of the technique to a wider range of chemical applications. In addition, the increased flux of high-energy neutrons available with the proposed pulsed

source will make possible experiments which are presently impractical with reactor sources. Generally speaking, an intense pulsed neutron source would be competitive with present high flux neutron sources for spectroscopic investigations for modes $\lesssim 1000 \text{ cm}^{-1}$. Such a source would provide distinct advantages for modes above 1000 cm^{-1} and for low Q studies (see below).

The OH stretching and bending regions ($\sim 1000\text{--}3000 \text{ cm}^{-1}$) of solid carboxylic acids contain a number of broad peaks. At present there are no quantitative explanations for the number and breadth of the peaks observed, though several theories have been proposed.⁶⁻⁸ Due to the unique properties of neutron scattering mentioned earlier, a study of this region would be invaluable in providing guidance and quantitative results for formulation and verification of theoretical explanations of this phenomenon. Due to the large energy transfer involved and the necessity of high resolution ($\sim 1\text{--}10 \text{ cm}^{-1}$) there is no other facility presently operating which could be used.

b. Instrumental Considerations

In studying the molecular vibrations of solids, a resolution of $\sim 10 \text{ cm}^{-1}$ and a Q of 3 to 10 \AA^{-1} is generally considered acceptable. For the low-Q investigations of fluids, Q should be $\lesssim 1.0 \text{ \AA}^{-1}$, and the energy resolution should be matched to the expected recoil broadening, which depends on the mass of the molecules being studied.

2. Small Q Scattering in Molecular Fluids

One of the chemical applications of a high intensity pulsed source would be in the study of molecular vibrations in gas or liquid phases. As originally demonstrated in the work of Brugger, et al.,⁹ the intrinsic "recoil broadening" of inelastic vibrational peaks makes it virtually impossible to measure discrete neutron spectra in fluid phases, unless the spectra are measured under conditions leading to low momentum transfer (high incident energy, low scattering angle). Unfortunately, these conditions result in a dramatic lowering of the inelastic cross sections (roughly proportional to Q^2) and often require well-resolved, copious monochromatic beams at high energies (100-500 meV). In fact there have been few, if any, successful measurements of vibration spectra in fluid phases by neutron scattering. The availability of intense neutron fluxes above 100 meV will thus open up the field of high resolution neutron spectroscopy in molecules. The primary applications of such spectral measurements would be twofold: (1) the observation of optically inactive or weakly active molecular vibrations and (2) the use of the vibration peak energies and the spectral profile (peak shapes and intensities) to obtain information about the nature of molecular force fields. This latter possibility of testing various molecular force models by direct calculation of neutron spectra for comparison with spectral data has been discussed at some length by Hudson, et al.,¹⁰ but as of yet has not received a completely satisfactory test due to the unavailability of high resolution neutron scattering results for "free" molecules. Other aspects of such calculations have already been mentioned in part 4.1 of this section.

In using an advanced pulsed neutron source to measure important molecular vibration frequencies, it is clear that a major application would be the study of large amplitude modes, most notably torsional vibrations in the $50\text{--}400 \text{ cm}^{-1}$ range. The low Q work of Grant, et al.,⁹ and others has shown the potential of

carefully chosen neutron experiments in this area, but almost all previous meaningful measurements have been done in solid phases. The selective application of this technique to prototype molecules (alkanes, substituted aromatics, etc.) has provided significant information on the nature of the potentials and nonbonded interactions governing internal rotation in such molecules. A particularly exciting prospect is the ability to measure higher torsional vibration states by low Q scattering. In fact, such measurements are essential to obtain quantitative information on the shape of the potential barrier. The small Q method could also be used to measure many other vibrational modes at moderate energy transfer ($< 800 \text{ cm}^{-1}$) including chain and ring deformation modes, bending modes and lower frequency torsional modes involving large-mass (high moment-of-inertia) segments.

It should be noted at this point that the high intensities available in the $1000\text{--}3000 \text{ cm}^{-1}$ range in an advanced pulsed source might allow the observation of molecular vibrations in low concentration ($\sim 1\%$) inert matrices, a technique which thus far has only been used in optical spectroscopy. The increased flux of epithermal neutrons would be important in making such a low contrast experiment feasible, while maintaining the capability of small Q measurements, thus providing (in principle) more well defined neutron spectra. Aside from the molecular systems mentioned above, such studies would be of considerable interest in strongly polar molecules. For example, a molecule of interest would be the acetic acid monomer whose vibrational assignments are still the subject of considerably controversy, in spite of extensive infrared studies.¹²

Finally, there are several general points which should be stressed with respect to the application of small Q neutron spectroscopy. First it would appear that, in spite of the very high intensity of epithermal neutrons, the practical range of measurements in fluid phases of most molecules will be $\lesssim 800 \text{ cm}^{-1}$. This is due to relatively broad absolute energy resolution ($\sim 20\text{--}40 \text{ cm}^{-1}$) which one must accept if one uses incident energies ($\sim 1\text{--}2 \text{ eV}$) necessary to reach the necessary Q ($\sim 1\text{--}2 \text{ \AA}^{-1}$). In addition, an excessively long flight path and large detector package would be required. Thus work on high frequency spectroscopy ($\gtrsim 1000 \text{ cm}^{-1}$) would for most practical purposes be restricted to studies in the solid phase at moderate to high Q values, Q ($\sim 4\text{--}10 \text{ \AA}^{-1}$). Secondly, it should be noted that it is many times desirable (particularly when seeking to study large amplitude vibrations in molecular crystals, hydrogen-bonded compounds and polymers) to use a low Q measurement in the $10\text{--}800 \text{ cm}^{-1}$ range to obtain an appropriately resolved spectrum. Thirdly, it should be stressed that successful application of small Q spectroscopy in gas or liquid phases will still require an appropriate choice of molecules, e.g., the chance of success will always be optimized if one focuses on the vibration, torsion, wagging motion, etc., of a low mass or moment of inertia group (OH, CH₃, NH₂, etc.) in a large mass (high momentum of inertia) molecule.

3. Electronic Excitations

a. Crystal Field Transitions

A growing number of chemists is interested in experiments to determine crystal field levels in the salts and coordination complexes of transition metal, rare earth and actinide ions. Where the materials are optically opaque, black or polycrystalline neutron incoherent scattering provides the only method. Progress has recently been reviewed¹³ and the subject is being

treated in greater depth in the magnetism panel. We therefore give no further details here.

b. Spectroscopy of Forbidden Singlet-Triplet Transitions

There is a large class of electronic transitions involving simultaneous electronic spin changes which are forbidden by optical selection rules, and are thus difficult to study directly by present techniques. Among these are the triplet-singlet transitions in O_2 and NO , and certain levels in transition metal ions. Because they are long-lived sources of energetically excited molecules, the triplet states of organic molecules are important intermediates in many chemical reactions, and it would be highly desirable to have a spectroscopic technique for their direct observation and characterization.

With a neutron source in the eV range these transitions could be studied directly via the interaction of the neutron magnetic moment with the molecular moment caused by the unpaired electrons in the triplet state. As outlined below, an order of magnitude estimate of the cross-section suggests that these transitions would be readily observable.

The interaction we consider is

$$\mathcal{H} = -\underline{\mu}_N \cdot \underline{H} ,$$

where μ_N is the magnetic moment of the neutron, and \underline{H} is the magnetic field caused by the spin and orbital magnetic moments of the electrons in the triplet state. For simplicity, we ignore the orbital magnetic moment. The molecular wavefunction $|\lambda\rangle$ is approximately separable into nuclear coordinates, electronic states, and spin variables; $|\lambda\rangle = |Q(\text{vib})\rangle |\phi(\text{electronic})\rangle |\chi(\text{spin})\rangle$. A very simple order of magnitude estimate of the cross-section for the singlet-triplet transitions can be made by approximating the singlet-triplet transition moment by the expectation value in the (magnetic) triplet state. In that approximation, the spin part of the total cross-section is identical with that of a paramagnetic ion,¹⁴ and the total cross section is a product of the spin portion and the vibrational part

$$\frac{d\sigma}{d\Omega} \approx \left\{ \frac{\gamma e^2}{m_e c^2} \right\} \frac{g}{2} |F(K)|^2 \frac{2}{3} S(S+1) \langle Q'(\text{vib}) | Q^0(\text{vib}) \rangle .$$

Here, the term in curly brackets is identical with that of a paramagnetic ion, $F(K)$ is the form factor for the electronic distribution of the unpaired electrons, and the vibrational overlap of the ground and excited state vibrational wavefunctions, $\langle Q' | Q^0 \rangle$, is the familiar Franck-Condon factor. A conservative typical value is $\sim 1/10$. The spin S is 1 for a triplet state.

The flux of IPNS II in the region of the singlet-triplet transition ($\sim 1\text{--}4$ eV) is of the order of the thermal flux of a steady state reactor such as CP-5 at Argonne, where magnetic scattering is readily observable. We thus expect that such transitions as the ground state singlet-excited triplet in

benzene should be well within the capability of IPNS II, opening up a new area of molecular electronic spectroscopy. Forbidden transitions in such other systems as transition metal ions should also be accessible. The only requirement is that either the ground or the excited state (or both) be paramagnetic.

c. Cooperative Electronic Excitations in Molecular Crystals

In addition to observing the density of states for electronic singlet to triplet excitations in molecular crystals it may be possible to study the exciton dispersion curves for triplet and singlet excitons as well as the exciton-phonon interactions in suitable cases. At present the only information on exciton dispersion curves is from observations of the Davydov splittings (for $Z \geq 2$) molecular crystals. These splittings occur in the polarized electronic absorption and fluorescence spectra at low temperatures. Strong phonon coupling is indicated in certain cases (anthracene second singlet system for example) by rather broad bands near the optical origins. For weaker transitions extensive phonon density of states spectra are built on the electronic origins and these have been used to study the exciton phonon coupling mechanism.

Following the arguments outlined in (b) we suggest that it may be possible to scatter neutrons coherently from electronic modes thereby building up the exciton dispersion curves directly. For typical molecular crystals the exciton bands lie in the 0.5 eV - 5 eV range and from the known Davydov splittings we estimate dispersions between 0.01 eV and 1 eV. The experiments would thus require energy resolutions within this range at the above energy transfers but would require momentum transfers Q in the range $0.1 \text{ \AA}^{-1} < Q < 5 \text{ \AA}^{-1}$ with a resolution of about $20\% = \Delta Q/Q$.

C. INTERMOLECULAR FORCES

1. Lattice Dynamics of Molecular Crystals

Aside from incoherent scattering studies of molecules and other complex compounds in solid and fluid phases which have been discussed above, a high intensity pulsed source would clearly be valuable in extending the application of coherent neutron scattering to studies of the dispersion curves of high frequency ($> 800 \text{ cm}^{-1}$) modes in molecular single crystals. Such applications would be much more limited in scope than the incoherent scattering studies discussed in the remainder of this report. These would include, e.g., the stretching and bending vibrations in strong H-bonded systems and intrachain bending made in polymers. Theoretical dispersion curves have been calculated for polyethylene which predict large dispersion in the chain direction for several high energy "intra-molecular modes ($E \sim 100 - 150 \text{ meV}$). Thus, a careful measurement of this dispersion could provide valuable information (in addition to dispersion curves measured for lower frequency modes) toward a detailed understanding of intramolecular forces in polymers. Similarly, measurements of dispersion curves and phonon lineshapes for high frequency modes in simple hydrogen-bonded crystals could provide important insights into intermolecular effects on hydrogen bonded crystals. It should be stressed here that such measurements are not especially suited to pulsed sources, and would obviously be limited by the availability of deuterated single crystals

of appropriate size of these materials unless the experimental conditions allow the use of polarization analysis for decreasing the incoherent contribution to the scattering.

The special applicability of this new pulsed source to lower frequency lattice dynamics studies, e.g., on low symmetry molecular crystals would depend on the development of a new generation spectrometer for collecting and sorting out a great number of phonon peaks over a wide range of reciprocal space and, perhaps more important, the analysis of such widely scattered data according to meaningful force models.

2. Quasielastic Scattering

Another class of experiments which could be done very effectively using a high resolution time-of-flight spectrometer ($E_0 < 20$ meV, $\Delta E \sim 0.01 - 0.02$ meV) at an advanced pulsed source is the study of the details of molecular reorientation in molecules and orientationally disordered crystals (organic plastic crystals, ammonium salts, crystal hydrates, etc.). Quasielastic neutron scattering has proven to be a powerful and in some cases unique probe of the geometry and rates of molecular reorientation over a wide time scale ($10^{-9} - 10^{-13}$ sec). Important information has also been provided on the dynamical mechanisms of order disorder phase transitions and orientationally disordered crystals.

It would seem that for such quasielastic scattering studies (over a Q range up to $\sim 6 \text{ \AA}^{-1}$) the proposed pulsed source would be competitive with the work on the highest-flux steady state sources. This technique is of course also widely applicable to the study of chain reorientation and other motions in solid polymers, to rotational and translational diffusion of molecules and polymers in solution, and of molecules on surfaces, trapped in layered structures, etc. A selection of these applications will be discussed in later sections in this report. For many of these systems, high resolution capability is necessary to fully unfold the various contributions to the scattering (for example, to separate translational and rotational diffusion modes in molecular liquids), and indeed at reactors line broadenings of $\pm 0.1 \text{ } \mu\text{eV}$ can now be routinely measured; this may prove a challenge to the ingenuity of instrument designers in fully utilizing the power of an IPNS type facility.

D. POLYMERS

1. Introduction

Neutron diffraction and inelastic scattering from polymers has led to considerable new insights for the structure and dynamics of these materials. Their intrinsic and commercial importance adds extra value to these results. Diffraction measurements have been concerned with understanding the disorder in polymer crystallites and studying polymer conformations by small angle scattering. Inelastic scattering studies have covered the first determination of phonon dispersion curves, the dynamics of side groups attached to polymer backbones, which are frequently important in mechanical and other relaxation processes, and studies of the quasielastic neutron scattering associated with random motions of the polymer in rubbery states. It seems likely that some

of these measurements can be extended with the higher fluxes available from the intense pulsed neutron source. For some others there will be a particular advantage in the pulse nature of the source. For the area of small angle scattering we foresee some difficulties in using the source because of the predominance of short wavelengths and the pulsed operation.

In the paragraphs which follow we review some of the possible applications of neutron scattering in the polymer domain and comment on the desirability of the proposed source for most work.

2. Structure and Disorder in Crystalline Polymers

Neutron diffraction from predominantly crystalline polymers has some advantages over x-ray methods especially when powder samples must be used. It is easy to obtain low temperatures and recent studies of annealing in crystalline materials indicate that quite high resolution powder patterns could be obtainable which could be processed by, for example, the Rietveld method.

In the last few years, also, it has been possible to obtain macroscopic specimens of fairly highly oriented polymers--often with 3-dimensional orientation.¹⁵ In conjunction with the measurement of phonon dispersion curves in such materials it is desirable to make studies of the neutron diffraction and it may be that the modified Laue method with multi-detector counters would be a valuable means for doing this.

3. Studies of Bulk Polymer Properties

It is of intrinsic interest to relate the bulk mechanical and other properties of polymers to their microscopic structure. Insofar as the intense pulsed neutron source diffractometers are well adapted to quick measurements of neutron diffraction from partly and highly oriented polymer specimens, it should be possible to use the methods of Sakurada¹⁶ to study the tensile modulus by observing the crystalline strain through shifts in the neutron diffraction peak positions after the imposition of an external stress.

For such studies, the pulsed nature of a neutron source might well prove to be a definite advantage since, with adequate computer support, the creep behavior after imposition of the external stress could be studied as a matter of course. Other applications where the pulse nature of the source is of value would be measurements of relaxation under temperature and pressure shocks (which are much easier to study than with x-rays because of the ease with which these shocks can be imposed in a neutron experiment) and swelling effects resulting from humidity and other changes in hydrophylic polymer materials.

4. Microscopic Properties of Polymers

In connection with the above measurements it seems possible that extensions to the work of the last decade in the field of the microscopic structure of polymers might be foreseen with the IPNS. We take first the most promising areas such as the extension of phonon dispersion curve measurements in the energy transfer region $1 \text{ cm}^{-1} < \Delta E < 3000 \text{ cm}^{-1}$, to high energy excitations

in polymers, studies of phase transitions in crystalline polymers by coherent inelastic scattering in the energy transfer region $10^{-2} \text{ cm}^{-1} < \Delta E < 500 \text{ cm}^{-1}$. In both of these areas there is much work to be done and the results lead to a direct understanding, at the microscopic level, of macroscopic mechanical modulus.

Another area of promise for the IPNS II is in the study of incoherent inelastic scattering from hydrogenous polymers. By this technique it has been shown that torsional oscillations of methyl side groups in polymers such as polypropylene are sensitive to the tacticity of the polymer.¹⁷ Also by incoherent inelastic scattering in the energy transfer range $10 \text{ cm}^{-1} < \Delta E < 500 \text{ cm}^{-1}$ it has been possible to show that the methyl torsion is sensitive to the configuration of block copolymers.¹⁸ On stretch oriented polymers measurements of the vibrational spectra and in particular the dependence of the vibrational intensities upon the orientation direction of the polymer with respect to the wavevector transfer might be able to give information about orientational relaxation effects in the polymer under stress. This would be a case where the time dependence of the neutron beam would be of value.

As far as we are able to see at the moment there are a number of areas where the present proposed source would be particularly valuable and where the results are of immediate interest to polymer scientists. The first of these concerns the determination of polymer conformation, radius of gyration, end-to-end distance, etc., for dilute solutions of, e.g., fully deuterated polyethylene in fully protonated polyethylene. Work of this kind has been pioneered by groups in France, Britain, and Germany with measurements at the Institute Laue-Langevin (Grenoble).^{19,20} Such measurements can be extended not only for polymers in the solid phase but for polymer solutions and there is a very considerable amount of work to do in this area. The results have already been most encouraging. As far as we can see the need to get to momentum transfers of the order of 10^{-2} \AA^{-1} for such measurements is rather difficult for the proposed spectrometer. Another area of some interest in polymers would be to study the conformations of adsorbed polymers and again small Q scattering measurements would be needed.

5. Biopolymers, Ionomers, Etc.

As in the measurements on synthetic macromolecules there are many studies to be done of the bulk modulus or elasticity for biopolymers. Collagen is a good example of this which is at present being studied at the Institute Laue-Langevin. Until now measurements on these materials--which are highly sensitive in their structure to the amount of water present--have not been made under controlled conditions of humidity, temperature, and pressure, etc. The whole domain of biophysical measurements exists and could readily be pursued, using the method of Sakurada [loc cit]. It also seems to us that a positive advantage of the pulsed source might well arise in these studies since, for example, in the studies of ion tractile and creep processes for biological materials the time dependence is often quite slow, or, in the case of muscle may be periodic under stimulation, etc. A limitation in the extension of this work is that most biological macromolecules have fairly long repeat distances. This means that again fairly low momentum transfers will need to be obtained. Typically we would think that the range might be $0.01 < Q < 5 \text{ \AA}^{-1}$.

There exists a domain of microscopic studies related to the macroscopic measurements above, some of which might be pursued with advantage with the pulsed source. Others because of the long period of repeat in the biopolymer are of less obvious attraction. From the point of view of elastic scattering we think that studies of the conformational transitions in oriented and partially oriented biopolymers as a function of pH and other variables could be studied to advantage in the region $0.1 < Q < 5 \text{ \AA}^{-1}$ which should be readily available. The small angle scattering counterpart of this would suffer from the same problems as those outlined above. Direct study of biopolymer structures, for example, using the contrast variation technique, have been already demonstrated for collagen, seem less attractive because they would have to be carried out in the momentum transfer region between 0.01 and 1 \AA^{-1} .

For the same reason studies of polymer dynamics in the macromolecular systems by inelastic neutron scattering seem to be severely limited because of the low energy transfers that would be needed [$10^{-2} < \Delta E < 100 \text{ cm}^{-1}$] and the associated low momentum transfers $0.01 < Q < 1 \text{ \AA}^{-1}$.²¹

6. Conclusions

This field of studies supports the request for an instrument to make diffraction measurements in the region $0.5 < Q < 10 \text{ \AA}^{-1}$ working with both powders and single crystals. In addition we would request the construction of an inelastic scattering spectrometer working in the region $10 \text{ cm}^{-1} < \Delta E < 4000 \text{ cm}^{-1}$ for the study of coherent inelastic scattering or incoherent inelastic scattering in this region. We would further request that detailed consideration be given to the problems raised in making small angle neutron scattering measurements in the region $0.01 \text{ \AA}^{-1} < Q < 1 \text{ \AA}^{-1}$.

E. SMALL PARTICLES, SURFACE, AND ADSORBED SPECIES

a. General

Surface science is an area of fundamental interest as well as high technological importance. The rapid development in the last decade of microscopic probes--mainly involving the scattering and emission of electrons--has yielded detailed information on surface reconstruction and chemisorption. Although neutrons are not usually a strongly interacting probe, they can in special cases be used to measure both structural and dynamical properties of surfaces and adsorbed species which are inaccessible by any other technique.

For example, within the past two years there have been several neutron diffraction investigations of the crystal structure and phase diagrams of atoms and molecules [N_2 ,²² H_2 ,²³ and ^{36}Ar ²⁴] adsorbed on (001) surfaces of graphite. In the case of ^{36}Ar the collective excitations within the film, and the atomic motions perpendicular to the substrate, have been determined. In addition, incoherent inelastic neutron scattering from hydrocarbon films has been used to study the effect of a carbon substrate on molecular vibrations and diffusion.²⁵

Since neutrons are not preferentially scattered from surfaces, only a few high specific-area substrates, and adsorbates with large neutron scattering cross sections can be studied at the present time. An increase of an order of magnitude in useful flux would open up this area to essentially all

adsorbates and a larger class of substrates. Interesting areas of study in relatively weakly bound (physisorbed) systems include: center-of-mass and orientational ordering in monolayers, epitaxial and non-epitaxial crystal formation and growth, phase transitions and critical phenomena in quasi-two-dimensional systems, collective excitations in films, molecular vibrations on surfaces, and surface diffusion.

Neutron sources with high epithermal neutron fluxes will enable study of the dynamical modes of chemisorbed species, even at very low coverages. This is a prerequisite for the study of heterogeneous catalysis which can involve "special" surface sites. At the present time, only one experiment has been reported on a chemisorbed system--hydrogen on Raney nickel (a commonly used catalyst).²⁶ In addition, measurement of structure factors and dynamical modes as a function of temperature would be of help in sorting out catalytic reaction pathways.

It is well established that particle size is an important factor in the effectiveness of certain catalysts. It is hoped that studies²⁷ of very low Q phonon modes in small ($\lesssim 100$ Å) particles will lead to a much needed increase in the understanding of the role of particle size in catalysis.

The nature of surface reactions and their eventual diffusion into the bulk is integrally related to the phenomenon of corrosion. With such an intense source, it may be possible to use neutron diffraction to make profiles in order to measure the extent of oxidation and of lattice strain as a function of time and distance from the surface.

b. Instrumental Considerations

To study both physisorption, chemisorption, and the dynamics of small particles by neutron scattering it requires the highest obtainable flux over a wide range of incident neutron energy. The operating range of a triple axis spectrometer (say at Brookhaven) should be extended to ~ 0.5 eV with a resolution of ~ 1 meV for the study of vibrations of chemisorbed species. At the same time one would like to retain the high resolution (0.02 meV) at low energy E_0 (~ 5 meV) currently available on the Brookhaven triple axis machines in order to study the lattice dynamics of small particles at low Q ($Q \gtrsim 0.1$ Å⁻¹, $\Delta Q \lesssim 10^{-2}$ Å⁻¹).

The study of surface structures, including superlattices, will require a $0.01 \rightarrow 10$ Å⁻¹ range of Q for the small-angle scattering spectrometer and $0.1 \rightarrow 50$ Å⁻¹ for the modified Laue spectrometer.

F. INTERCALATION, ABSORBED MOLECULES AND NONSTOICHIOMETRIC COMPOUNDS

1. Introduction

Operation of industrial catalysis (of heterogeneous variety) is not only concerned with the dynamics and structure of molecules adsorbed on surfaces (discussed in the last section) but does concern the partial or total absorption of molecules or atoms in some types of catalytic materials. And so it is relevant here to consider what benefits may be obtained from the intense pulsed neutron source for studies of intercalation, absorbed species, and

other nonstoichiometric materials. There is a wide variety of possibilities in this class of materials which includes zeolites and molecular sieves, layer structures such as graphite, tantalum disulfide and clay minerals as well as clathrates and hydrides. These materials are commercially exploited as catalysts but also show some promise (for example, TaS_2 compounds) as potential high temperature superconductors. In these materials the intercalated or adsorbed molecules frequently have a variety of different structures. The structures may be orderly or there may be disorder arising from molecular reorientation or from molecular conformational changes. The study of this disorder is of intrinsic interest and may be of practical value.

Measurements with neutrons have been made on most of the classes of materials referred to above using both neutron diffraction and using neutron inelastic scattering. We review the likely advantages of these two methods with examples below.

2. Diffraction Studies

a. Ordered Phases

As examples of studies in the structure of ordered, absorbed, and intercalated species we consider the work that has recently been done on hydrides²⁸ and the dichalcogenide compounds and Lewis bases such as ammonia.²⁹ In the hydrides diffraction information, e.g., on PrH_x has shown how, as a function of the absorbed hydrogen concentration the fraction of octahedral and tetrahedral sites are filled in this crystal. For the $\text{TaS}_2\text{-NH}_3$ compound powder diffraction studies on both the adsorbed in ND_3 and NH_3 systems are of sufficient quality to allow recent models for the siting of the NH_3 ²⁹ to be tested. It seems likely to us that such studies may be immediately extended both with powder diffraction and with single crystal diffraction to a wide variety of similar intercalation compounds of potential interest for high temperature superconductor studies.

b. Disordered Materials

If we take the stoichiometrically filled $\text{TaS}_2\text{-NH}_3$ compound at a low temperature the crystal structure as revealed for example by the powder diffraction, indicates ordering of the center of mass of the ammonia molecule and possibly also of the hydrogens with respect to the crystal c axis. As the temperature is raised this order breaks down due to rotatory motions of the molecule and this is immediately apparent in the diffraction pattern. Such systems are interesting models for other phase transitions and it is of intrinsic interest to study them from a number of points of view. It is clear that the powder diffraction possibilities with the intense pulsed neutron source make such studies with it very fruitful.

In addition to disorder in the stoichiometric materials there may be disorder of both translational and rotational kinds when the material is incompletely filled with the intercalating molecule. For substances where this situation arises neutron diffraction measurements may prove valuable for finding preferential sites for the intercalated or adsorbed molecules. This is important in layer compounds but also in materials like zeolites and

molecular sieves of considerable importance commercially from a catalytic point of view. These substances frequently have two or more sites within the aluminosilicate cages and neutron diffraction measurements to study the localization of molecules and molecular fragments on these sites, possibly during reactions, is of considerable interest in studying the catalytic process.

In addition to the disorder which arises from rotational motions of molecules in the stoichiometric materials from noncomplete filling, and hence translational disorder, it is possible in many layer structures to have several polytypes in the basic chalcogenide crystal. Their presence together in microscopic crystals makes studies with those large specimens extremely difficult unless particular care has been taken to get a single polytype species. Such problems are not so acute, it appears, in powder studies where single polytype specimens of small dimensions can be fairly easily prepared. Hence, it is quite likely that many studies will be made with powder specimens and again the virtues of the powder diffraction apparatus on the intense neutron beam source are apparent.

c. Diffusion and Kinetics

The formation of intercalated materials, a process of diffusion in of the intercalating molecule, can be studied by observing the change in the Bragg pattern, for example, from powder specimens, as a function of time. Pioneering work along these lines has been done by C. Riekel (ILL-Grenoble) in studying the tantalum disulfide-ammonia system. It is clear that for studying diffusion and the kinetics of exchange, for example in such systems, the pulsed nature of the IPNS, for example, would be of considerable attraction. It is clear once again that such measurements can be extended to a wide variety of systems which may embrace not only diffusion of the molecules into the lattice but also conformational changes of molecules once adsorbed. For example, many layered structures such as the micas intercalate long-chained hydrocarbon amine and as a function of temperature the amine conformation changes in a discontinuous way due to geometrical isomerization.³⁰ To study such phase transitions neutron elastic scattering has some advantages because of the possibility of isotopic substitution. More importantly, such studies would be presupposed by an inelastic scattering measurements to study the dynamics of such systems.

3. Inelastic Scattering

Neutron inelastic scattering has a number of advantages over optical, nuclear magnetic resonance, and other methods of spectroscopy for studying the motions of absorbed molecules or intercalated materials which are either completely absorbing or reflecting to light. This arises because frequently the intercalating material, e.g., graphite, alumino-silicate, has a low scattering cross section compared to the scattering cross section for the included molecules. In the tantalum sulfide-ammonia compound it has been possible to observe quasielastic neutron scattering associated with translational and rotational modes.³¹ These measurements have also made it possible, using Be-filter spectroscopy,³² to reveal the torsional oscillations of the ammonia trapped between the layers in this system.³³ These measurements make use of the high incoherent scattering from the ammonia included between the

layers and the large amplitude of motion associated with the torsional vibrations. Many other examples of these types of measurements could be given, but it is clear that for studying molecular translational and rotational diffusion within intercalated species neutron scattering offers important complementary information to nuclear magnetic resonance studies where these can be made, and where these cannot be made it gives a unique method of studying such motions.

It seems to us that the most obvious possibilities with the intense pulsed neutron source will be to study energy transfers in the region of about 1 cm^{-1} to 10^3 cm^{-1} , in which case the molecular vibrations and torsions might well be observable in such systems. If spectrometers, working above 10^3 cm^{-1} , can be made then measurements of other molecular vibrational modes especially in the ordered phases would be interesting. Such measurements would be complementary to infrared studies, which are possible for such materials as zeolites only within the small windows for the electromagnetic spectrum where the host lattice is not strongly absorbing. It appears to us that considerable work in this field is yet to be done.

As to studies of quasielastic scattering ($10^{-3} \text{ cm}^{-1} < \Delta E < 1 \text{ cm}^{-1}$) our first impressions are that IPNS will be less advantageous than conventional reactors. If instruments such as the backscattering spectrometer or the spin-echo machine can be envisaged, then interest in this area would be extremely great because the extremely high resolution is necessary for the proper separation of translational and rotational diffusion processes.

4. Conclusions

We conclude that the area of intercalated, absorbed, and nonstoichiometric compounds is of considerable intrinsic interest from the point of view of diffraction studies of the structures of these materials and from the point of view of the kinetics of their formation and the dynamics of molecules intercalated. These studies, using neutrons, are as yet at a very early stage and the pulsed nature of the source as well as the good spectrum of neutrons at short wavelengths have unique advantages for this field.

G. MOLECULAR LIQUIDS, SOLUTIONS, AND CHEMICAL REACTIONS

Considering the present state of theoretical development we feel that the basic problems associated with liquids and solutions in general are largely those concerned with the statistical mechanical description of a complicated many-body system in terms of the fundamental interactions between the constituent fluid molecules. We foresee two types of problems as being relevant at this time. First, the study of the equilibrium structure of molecular fluids is receiving a great deal of theoretical attention, and correspondingly there will be much experimental interest (see the discussions of the Liquids panel).³⁴ It may be useful to note here that the region of very large momentum transfer ($k > 30 \text{ \AA}^{-1}$) will constitute a unique capability of IPNS in probing the shape of molecules (bond distances and angles) in the liquid and gas phases.³⁵

The second type of problems involves the dynamical structure of molecular fluids. In the case of one-component liquids one should study the dynamics of reorientations and translational diffusion whereas for two-component fluids one should initially focus on simple monatomic systems. This point of view is necessitated by the fact that scattering spectra of this type of measurements are not useful unless quantitative theoretical analysis can be carried out.

The approach in which the properties of solid and liquid phases of a molecular system are assumed to be described by the same set of interatomic interaction potentials is considered to be generally useful. This implies that the structural and dynamical properties which are characteristic of the liquid phase can be regarded as manifestations of the statistical behavior of the system. It follows that if the molecular forces are already well known, neutron scattering data can be used to test the current theoretical methods for calculating the properties of a many-particle system. However, if the forces are not well established, then the neutron data can be used in conjunction with the other experimental information to determine the parameters of the assumed potential functions. In either case one is faced with the statistical mechanical problem of calculating observable microscopic and macroscopic quantities using potential functions.

In the case of molecular solids the method of lattice dynamics is quite well developed for computing the vibrational properties in terms of potentials. For molecular liquids the corresponding technique appears to be computer molecular dynamics. Given the potential functions this technique can generate all the quantities of interest for neutron scattering studies under well controlled conditions. While the computer calculations are useful in interpreting neutron scattering data, ultimately one would want to understand the computer and neutron results in terms of a dynamical theory of liquids. Current developments in kinetic theory of dense fluids may provide the basis for this kind of understanding.

Given the possibility of measuring the functions $S(Q, \omega)$ and $S_s(Q, \omega)$ to 1% accuracy, the problems in the following areas would be of considerable interest:

1. rotation-translation coupling effects
2. temperature and pressure dependence of relaxation times for molecular reorientations
3. self and mutual diffusion in two-component fluids
4. wavenumber dependent concentration fluctuations in two component fluids.

Kinetic theory provides the basis for a quantitative treatment at the molecular level of all the effects mentioned. In this approach collisions are treated explicitly in terms of intermolecular potentials and the density dependence is explicit. Through kinetic theory one can correlate the high-frequency, short-wavelength fluctuations in neutron scattering with the low-frequency, long-wavelength fluctuations described by transport coefficients.

We consider three applications to illustrate the kinds of problems one can study in one- and two-component fluids and chemically reacting fluids. In a hydrogen-bonded liquid incoherent scattering measurements in the intermediate Q region may show the dynamics of cooperative proton exchange known to exist in case of water.³⁶ In a binary mixture of hydrogenous and non-hydrogenous liquids one can measure $S_{HH}(Q, \omega)$ by incoherent scattering. Considerations based on kinetic theory show that the lineshape of the quasi-elastic peak is determined by a certain combination of the self-diffusion and mutual diffusion coefficients in the limit of small Q . Lastly, incoherent scattering measurements on a chemically reacting hydrogenous liquid provides an interesting opportunity of studying the wavenumber dependence of proton concentration fluctuations. Specifically Lassegues and White³⁷ have studied the proton transfer reaction between hydronium and trifluoroacetate ions in water solutions. The interpretation of the results is not yet clear because of low-resolution instruments used in the investigation.

Many of the scattering experiments envisaged here will be concerned with the low-frequency (or quasielastic) region of the spectrum. Reasonably good energy resolutions (~ 1 meV) and small Q (~ 0.01 and up) are required. Since these are not severe requirements, the experiments can be performed with either a pulsed or a dc source. However, there are important exceptions. These are the large- Q structure factor measurements and the measurements on chemically reacting systems which require small Q and energy resolutions of order 10^{-4} meV.

H. BIOPHYSICAL CHEMISTRY

One of the potentially powerful uses of neutron scattering techniques in biophysics arises from the high sensitivity to the dynamics of hydrogen atoms. For example, selective deuteration of a protein such as hemoglobin or myoglobin would enable one to "mask" that portion. The width of the quasi-elastic peak at small Q ($< 0.1 \text{ \AA}^2$) and high resolution will probe mainly the translational motion of the hydrogenated portion of the molecule, while that a higher Q probes mainly rotational diffusion.³⁸ It would be valuable to study the motional changes in such systems, as a function of the binding to O_2 , CO, etc. Similar studies might also be done on appropriate enzymes. Because of the necessity of working in dilute (1-5%) solution, high sensitivity would be required for these experiments.

Because of the pulsed nature of the IPNS source, it may be possible to make real-time measurements of the change in structure of membrane model systems as ions and other solutes diffuse into them. Other valuable real-time studies would include stress relaxation in collagen fibers, muscle contraction ($\tau \sim 10^{-2} - 10^{-3}$ sec),³⁹ and the dynamics of the response of liquid crystalline phases to applied electric and magnetic fields ($\tau \sim 10^{-2} - 10^{-5}$ sec). For time scales long compared to the neutron pulse separation ($\tau > 1/60$ sec), one can directly follow the response, at each pulse cycle, from an applied step function in the external field. Such an experiment has recently been reported on ferroelectric relaxation in NaNO_2 ,⁴⁰ and would be useful to study the longer time instabilities in liquid crystals (e.g., dynamic scattering). For shorter time processes, it would be necessary to phase the electric field pulses relative to the neutron source and use phase sensitive detection.

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SUMMARY CHART I

<u>Section</u>	<u>Research Area</u>	<u>Machine (Classical)</u>	<u>Range (cm⁻¹)</u>	<u>Resolution ΔE (cm⁻¹)</u>
MOLECULAR EXCITATIONS				
B.1	Molecular Vibrations in Solids Torsions H-Bonds	TOF, ¹ Be Filter	10-4000 1-500 10-4000	1 → 10
B.2	Small Q Scattering by Molecular Fluids Crystal Fields Electronic States	"Small K"	10-1000 1-50,000 100-50,000	10=1000
C	Intermolecular Forces and Excitations H-Bonds Plastic Crystals	3 axis, ² *TOF ¹	100 → 100-4000 0.1-1000	1 → 10 10 as good as possible
D	Polymers (a) Elastic Scattering (b) Inelastic Scattering	Low Angle Scattering ³	10 ⁻³ -3000	8 × 10 ⁻³ ; 10
E	Small Particles, Surfaces, and Adsorbed Species (a) Elastic (b) Inelastic	Powder, 4 circle Modified Laue ⁴	160 × 10 ⁻³ -4000	10 ⁻² ; 10
F	Intercalation, Absorbed Species and Nonstoichiometric Compounds (a) Elastic (b) Inelastic	Powder Diffraction	8 × 10 ⁻³ -4000 8 × 10 ⁻³ -4000	8 × 10 ⁻³ ; 10 8 × 10 ⁻³ ; 10
G	Molecular Liquids, Solutions, and Reactions (a) Elastic (b) Inelastic	Liquids Diffractometer "Small K", Backscattering	1-4000 10 ⁻⁴ -10 ²	10 ⁻⁴ ; 1

Summary Chart I (cont.)

<u>Section</u>	<u>Research Area</u>	<u>Machine (Classical)</u>	<u>Range (cm⁻¹)</u>	<u>Resolution ΔE</u>
H	Biophysical Chemistry Liquid Crystals, Transient Phenomena (see also Biophysics)		1-200	

SUMMARY CHART II

Section	Research Area	Range of (\AA^{-1})	Wavevector Resolution ΔQ	Uses Pulses to Advantage	Uses High E, High Q	Score and Comments		
						A [†]	B	C
B.1	Molecular Vibrations in Solids	0.1-10	0.1	✓	✓	A	(E > 100 meV) [B(E < 100 meV)]	
	Torsions	0.1-5			✓	A		
	H-Bonds			✓	✓	A		
B.2	Small Q Scattering by Molecular Fluids	0.1-2 \AA^{-1}				A		
	Crystal Fields	0.01-1.0	0.02	✓	✓	A		
	Electronic States	0.01-3		✓	✓	A		
C	Intermolecular Forces and Excitations	0.1-5	0.1	✓				
	Quasielastic Scattering	0.1-5	0.1 → 5		✓		B	
	Single Crystal Lattice Dynamics	0.1-5	0.1 → 5				B	C
D	Polymers			✓	✓			
	(a) Elastic Scattering	0.001-1	10 ⁻³			A (high Q)	[C (low Q)]	
	(b) Inelastic Scattering	0.01-5	10 ⁻²				B	[C (low ΔE)]
E	Small Particles, Surfaces, and Adsorbed Species	0.01-10	10 ⁻²	✓			B (high Q; kinetics of adsorption studies)	
	(a) Elastic	0.1-50			✓	A		
	(b) Inelastic	0.05-5	10 ⁻²	✓	✓	A (high E)		
F	Intercalation, Adsorbed Species and Nonstoichiometric Compounds			✓	✓	A (kinetic studies)		
	(a) Elastic	0.1 → 10	10 ⁻² → 1			A	B	
	(b) Inelastic	0.1 → 10	10 ⁻² → 5	✓	✓	A		

Summary Chart II (cont.)

Report Chapter	Research Area	Range of Q (\AA^{-1})	Wavevector Resolution Q	Uses Pulses to Advantage	Uses High E, High Q	Score and Comments		
						A [†]	B	C
G	Molecular Liquids, Solutions, and Reactions							
	(a) Elastic	1-50	$1.0 \rightarrow 10$				B	
	(b) Inelastic		5×10^{-2}					C
								(quasielastic)
H	Biophysical Chemistry, Liquid Crystals, Transient Phenomena (see also Biophysics)	0.01-10	10^{-3}	✓			B	

Summary Chart Rating Scheme

Instrument developments needed to achieve score given

1. Large aperture choppers of high speed
2. (a) Programmable multiscaler, e.g., to rate constant Q scans
(b) Time resolving multiscaler with very fast encoding
3. Very low angles
4. Needs time resolving area detector

[†] Meaning of the scores

A = We can identify important scientific uses which are impossible or incompletely done by the present steady state sources.

B = Competitive with present steady state systems.

C = Less appropriate applications.

III. REPORT OF THE PANEL ON CHEMICAL STRUCTURES OF CRYSTALLINE SOLIDS AT THE WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. INTRODUCTION

Inelastic diffraction atoms act as point scatterers and neutrons therefore have a clear advantage over x-rays in such experiments provided they are available with adequate intensity. Many applications of neutron diffraction have been limited or impractical because of intensity limitations and the related problems of obtaining adequately large samples and sufficient resolution.

Much of the work described below would become feasible through an increase in intensity and, especially in the area of powder diffraction, through the increased resolution made possible in a pulsed-neutron source such as proposed here. Furthermore, the substitution of time resolution for space resolution in time-of-flight studies facilitates the installation of special equipment, such as high pressure cells, bulky magnets and small window low and high temperature equipment. The elastic scattering experiments described in this section exploit further specific advantages of neutron diffraction like detection of hydrogen, sensitivity to hydrogen/deuterium ratio and isotope composition in general, sensitivity to orientation of the spins of magnetic nuclei and ability to detect very light atoms in heavy atom metal organic complexes.

The single crystal experiments proposed are subdivided into materials science, crystallography and fundamental physical and chemical properties, through these classifications obviously overlap and are therefore only loosely defined. Powder diffraction experiments present a wide field of new applications and are considered separately in a following section. Experimental considerations are considered in the second half of the section, including proposed instrumentation and requirements on the reproducibility of machine characteristics such as pulse height and shape and their long-term stability at all wavelengths.

B. PROPOSED SINGLE CRYSTAL STUDIES

1. Materials Research Applications

a. Superconducting Materials and Metal-Insulator Transitions

The ability to apply pressure and to achieve very low temperatures are of considerable advantage in studies of superconducting materials. β tungsten type superconducting alloys for example undergo a phase transition from cubic to tetragonal on cooling,¹ which will be accompanied by twinning unless an anisotropic pressure is applied to the sample. Thus, low temperature single crystal studies of these materials require simultaneous cooling and application of pressure. Such twinning is a quite general phenomenon in phase transitions in alloys in which the symmetry is lowered and its prevention makes a new range of measurements feasible. A second application is the study of metal-insulator transitions in one dimensional conductors such as KCP and TTF-TCNQ type complexes. Availability of high intensity would allow detailed measurement of the weak satellite reflections below the temperature of the Peierls transition and lead to knowledge of the actual atomic displacements accompanying the distortion. Very low temperature superconducting transitions as occur in polymeric sulfurnitride $(SN)_x^2$ are presently not accessible by

single-crystal diffraction. Use of a pulsed source with its implied reduction in required crystal motion during data collection enables the study of such phase transitions in this class of potentially very important materials.

b. Catalysts

An intense pulsed neutron source offers two major advantages for the study of catalytic systems. First, neutrons allow precise examination of hydrogen and other light atoms (positions and mean-square amplitudes of thermal motion) in the presence of heavy elements. Many catalysts (e.g., uranium alkyls and other organometallics) contain heavy metals. The study of hydrogen-atom position, and possibly hydrogen/deuterium exchange in such systems is of considerable importance. Second, an intense source would allow the use of small samples, which could open up neutron single crystal studies of zeolites, and other catalytic materials of which large single crystals cannot be grown. Thus far some x-ray work has been done on zeolite systems with promising results,³ but it has not been possible to locate all of the light atoms in these compounds at low temperature. If suitable single crystals could not be obtained, high resolution time-of-flight powder diffractometry could provide useful data for zeolites and other systems of importance in catalysis. Finally, the use of neutron sources to probe chemisorbed and physisorbed substrates on catalytic surfaces which have been treated in the section on chemical spectroscopy should be mentioned.

c. Metal Hydrides

A variety of alloys and metals (e.g., the intermetallic compound FeTi)⁴ have been tested as hydrogen-storage media. Large amounts of hydrogen can be added to such systems and the resulting hydrides are readily decomposed, but the detailed nature of binding of hydrogen in these hydrides is unknown. Neutron diffraction offers the only possible means of determining the location of hydrogen in these compounds, and such studies have been begun at the Brookhaven High-Flux Reactor. It would be highly desirable to carry out such studies at a variety of temperatures and pressures, and time-of-flight powder diffractometry would be ideal for this purpose. In addition, neutron incoherent scattering can be used to provide diffusion constants and other transport properties in such systems.

Recently, a variety of novel complex transition metal hydrides have been synthesized, and x-ray and neutron diffraction studies of such systems have yielded information on their structure. While it is sometimes possible to qualitatively fix hydrogen-atom positions in such systems, precise and positive location of protons requires the use of neutron diffraction. Many interesting compounds in this class of hydrides are extremely difficult to crystallize and the possibility to use very small single crystals or powders would greatly enhance this field.

d. Special Conditions

The use of time-of-flight techniques allows one to explore large areas of reciprocal space with detectors placed in a small number of fixed positions. Such is the case for diffraction both from powder and single-crystal samples, although in the case of single crystals, the number of required crystal settings to explore a given portion of the reciprocal lattice is inversely related to the number of detectors. Therefore, time-of-flight techniques are particularly advantageous for the study of samples under special conditions of pressure, temperature or magnetic field, where a high-pressure cell or oven, for example, may restrict access to the sample. A whole host of applications of the time-of-flight technique to the study of structural or magnetic phase transitions may be envisioned. Increased flux would be a particular advantage in high-pressure studies where it would be possible to maintain only small samples at the desired pressures. Also, quantities such as the magnetic field or pressure could be varied in a manner synchronized with the pulsed source, potentially allowing the study of relaxation phenomena.

2. Crystallographic Applications

a. Study of Extinction

Present theories predict a specific wavelength dependence of extinction. Theories for both primary and secondary extinction have been improved in recent years and represent extinction reasonably well unless the effect of extinction is severe.⁵ Nevertheless two major shortcomings remain. First, the treatment is based on the mosaic crystal model rather than on a more realistic description based on crystal dislocations. Second, and related the effect of diffraction in the mosaic blocks is described by intensity rather than by amplitude coupling. An improved model being considered at present would involve a continuous transition between the dynamical and kinematical treatments allowing for partial coherence for small misorientation in adjacent parts of the sample.⁶ One of the optimal ways of testing such theories is through the predicted wavelength dependence and this can be done much more easily and more comprehensively with a white beam and a time-of-flight diffractometer.

b. Anomalous Dispersion

Certain nuclei (e.g., Sm^{149} , Cd^{113}) exhibit resonances in the thermal region, with the accompaniment of anomalous dispersion. These anomalous effects are an order of magnitude larger than those observed in x-ray diffraction, and are markedly wavelength dependent. Neutron anomalous dispersion effects have been used successfully at high-flux reactors to determine phases for a cadmium derivative of myoglobin⁷ and a samarium EDTA complex.⁸ However, the measuring times needed to produce the required accuracy of counting statistics were quite long, largely because of the need to go to short wavelengths ($\lambda < 0.9 \text{ \AA}$) for which intensities fall off quite dramatically at present high flux reactors.

It should be noted that use of a hot moderator at ILL has considerably increased available intensities in this short wavelength region, compared with the two U.S. high-flux reactors. With the proposed IPNS, far larger flux would be available in this region than at ILL, and this, coupled with the

advantage of simultaneous measurement at many wavelengths with a time-of-flight Laue technique, would lead to increased accuracy and shorter counting times in anomalous dispersion measurements. However, the requirement that each reflection be measured at a minimum of two and preferably three or more wavelengths, would dictate that more counters be used than the minimum required for single-crystal work.

3. Fundamental Physics and Chemistry

a. Charge Density in Solids

The determination of charge densities in solids requires unbiased positional and thermal atomic parameters that can be obtained most favorably with neutron diffraction. Here accuracy is of crucial importance as the errors in the deformation of the experimental charge density on bond formation depends directly on the accuracy of the atomic parameters. Availability of a high-intensity neutron source would reduce uncertainty due to counting statistics and/or allow measurements on a smaller sample crystal with the concomitant reduction in extinction, absorption and multiple-reflection effects. At least equally important, high intensity and convenient cooling equipment allows the measurement of charge densities as a function of transport properties in materials with temperature-dependent conduction. Such measurements in conjunction with density-of-state measurements with spectroscopic techniques may become of tremendous importance in our understanding of these solid-state properties. For example, a charge transfer of 0.6 ± 0.15 electrons from TTF to TCNQ at 100 K was determined by diffraction.⁹ Measurement of the temperature dependence and the spatial variation of this charge transfer within each molecule would significantly enhance the understanding of the transitions observed at 58, 54, and 38 K.

b. Study of Molecules in Their Excited States

The pulsed nature of the source opens the possibility of a time resolved experiment in which an external perturbation such as a high-intensity laser source or a strong magnetic field is synchronized with the neutron source. The pulsed neutron source combined with a pulsed laser permits much higher light intensity than attainable on a steady-state experiment. In practice the time resolution will be translated into a wavelength resolution as the counter receives a practically continuous current of neutrons, which have passed through the sample at different times depending upon their wavelength. Such an experiment allows study of excited states provided a sufficiently high percentage of the molecules can be excited by laser pumping to allow coherent diffraction (otherwise the signal becomes part of the diffuse scattering pattern). In general, crystal structure may break down when an appreciable percentage of the molecules are in their electronically excited state but several interesting exceptions may be predicted. Some of these involve intramolecular hydrogen transfer in anils of salicylaldehyde¹⁰ which has a relatively small effect on the shape of the molecules and therefore on their packing. More generally crystals may also be vibrationally excited by selective excitation of their normal modes thus leading to a direct determination of the displacement coordinates through analysis of anisotropic Debye-Waller factors.

c. Organic Molecules

Single-crystal neutron diffraction studies at reactors have provided a wealth of precision data on organic structures. Neutron diffraction is the only technique for the determination of accurate hydrogen-atom positions in complex crystals. Since interactions among hydrogen atoms dominate non-bonded interactions in organic systems, such studies are important in providing data for conformational energy calculations and of interatomic potentials. With IPNS-II, such studies might be extended to materials for which large enough crystals for use on steady-state sources might not be available. It would also be possible to do accurate powder studies on fairly complicated systems, possibly with as many as 100 atoms per asymmetric unit.

d. Hydrogen Bonding

The IPNS would allow the study of hydrogen-bonded systems under conditions of temperature and pressure not accessible with conventional sources. These studies would also include investigations of H/D distribution as a function of temperature in systems with more than one type of hydrogen bond. Previous studies have shown that in such mixed systems protons accumulate preferentially in the stronger hydrogen bonds and a study of isotope distribution therefore gives a measurement of bond strength. It is not known whether the equilibrium is temperature dependent or established at the time of crystallization. Studies of hydrogen-bonded ferroelectrics and ferroelastics as a function of temperature may also be mentioned.

In general, the IPNS would allow extension of the work with conventional neutron sources, which has contributed tremendously to our knowledge of hydrogen bonding in solids.¹¹

C. PROPOSED POWDER STUDIES

The proposed pulsed spallation neutron source is capable of producing a significant increase in the potential of neutron powder diffraction. We shall outline the characteristics of the three proposed powder diffractometers, the high-resolution, the high-pressure and the high intensity instruments. A more detailed description of these instruments is given in the section on instruments. First, the high resolution diffractometer (HRD) has a multiple counter assembly with the counter at the diffraction angle $\theta = 88^\circ$, giving a resolution of $\Delta d/d = 0.001$ for the range $0.8 \leq d \leq 1.466$ Å. This represents a ten-fold improvement in resolution over the best available constant wavelength instruments. In addition, the intensity of the IPNS II source in this application is considerably higher than that of the highest flux instrument at ILL.

A second instrument, the high-pressure diffractometer (HPD) currently envisioned consists of a cubic press with detectors at $2\theta = 90^\circ$ covering a 90° acceptance angle. The resolution would be $\Delta d/d = 0.025$ for the range $0.5 \leq d \leq 5.6$ Å. Again this would represent a significant improvement in resolution and we expect a similar ten-fold increase in intensity making the use of this type of pressure system possible.

Third, the high-intensity small-sample diffractometer (HID) although at moderate resolution would be capable of high data collection rates for samples 0.1 - 10 milligrams. Use of a dual counter array at $2\theta = 90^\circ$ and 30° combined with time focusing will give a resolution of $\Delta d/d \sim 0.01$ which is comparable to the best constant wavelength instruments. Clearly then these three instruments represent a vast improvement over the best currently operating powder diffractometers. In a consideration of the possible applications of these instruments we shall consider the current work on existing instruments and then attempt to extrapolate given the apparent increases in both resolution and intensity.

In the area of structural studies the use of profile fitting least-squares¹² has enabled one to analyze complex constant wavelength neutron diffraction¹³ patterns with as many as 800 Bragg reflections contributing to the profile and refining 50-60 variable structural parameters. In these profiles much of the high Q portion of the diffraction pattern had more than 40 reflections contributing to each point in the pattern. For the proposed HRD instrument the ten-fold improvement in resolution would mean nearly complete resolution of these patterns with only 3-6 reflections contributing to each point. In both of these cases we consider a reflection to make some contribution over a range that is three times the full width at half maximum. With this extremely high resolution it would be possible to assign relatively accurate structure factors to as many as 1000 reflections in a powder pattern, which would enable the solution of unknown structures with as many as 20 unique atoms. These structures could then be refined to high precision with the profile-fitting technique. If 40 reflections contributing to each point in the diffraction profile is considered to be a limitation (it may be much larger than this) then a diffraction pattern made up of 1000 reflections could be analyzed by least-squares profile-fitting with 200-300 parameters (or more). This is an extremely powerful structure determining tool which could handle nearly all small and medium size crystal structure analyses currently done by single crystal techniques. It is clear, however, that to realize these improvements, the characteristic line shapes and their dependence on wavelength must be known with great precision.

A second aspect of this HRD is that the high intensity would permit a combination of smaller samples and shorter counting times for less complex crystals. This would permit the use of the HRD and especially the HID for rapid examination of many analytical samples. In addition, the investigation of surface structure, which is inherently a small sample, could also be feasible, particularly if the surface is composed of strongly diffracting molecules such as hydrocarbons or nitrogen oxides on a weakly diffracting substrate. This kind of study could increase our knowledge of catalytic surfaces and mechanisms and avoid the difficulties of interpretation that accompany LEED experiments. The high flux also permits the investigation of small samples. In particular, compounds of most of the synthetic actinide elements can only be obtained in milligram quantities and cannot be studied by any other means.

Since the neutron beam is pulsed, TOF diffractometry permits a fixed scattering angle. This is especially suited to high pressure diffractometry where access to the sample is highly restricted by the pressure equipment. Because of the flux and resolution of current instruments only relatively simple substances have been studied at high pressure.¹⁴ With the higher

resolution and higher intensity of the HPD a more complex structures can be studied at pressure particularly if the analysis of the pattern is done by a profile-fitting technique. Examples of new work that can be performed with this facility include studies of geochemical reactions at the conditions existing in the earth's mantle which can aid in understanding of the processes of mineral formation and behavior.

The extremely high resolution in the HRD gives an instrumental line width that is comparable to that due to typical particle size, thus permitting an investigation of line broadening by anisotropic particle shapes. This line broadening can also arise from preferential strain in polycrystalline materials. Investigation by high-resolution neutron diffraction of such materials under a uniaxial stress can lead to an insight into the strength of materials. Because of the penetrating power of neutrons, much larger specimens than those amenable to x-ray studies can be used. In particular, ceramic materials are especially suited for such studies.

The high resolution also permits the investigation of very small lattice distortions due to ferroelectric and antiferroelectric transitions. Recently Hewat¹⁵ investigated these transitions in perovskites and was able to detect changes in the lattice parameters to a precision of $\pm 0.001 \text{ \AA}$ and $\pm 0.2^\circ$ via a profile fitting analysis. He obtained similar results in an investigation of $\text{NH}_4\text{H}_2\text{PO}_4$ and $\text{NH}_4\text{H}_2\text{AsO}_4$. It is clear that both the HRD and the HPD will be useful in these studies particularly at high pressures where only TOF techniques can be used.

The pulsed nature of the neutron source permits the study of transient effects by the use of a synchronized external field (electric, magnetic or pressure). Recently Niimura and Muto¹⁶ have demonstrated this type of study with the example of the ferroelectric transition in NaNO_2 . An external rectangular step pulse of high voltage was applied synchronously with the Tohoku University Linac neutron pulse. It was concluded that the NO_2 group rotates about the (001) axis when the direction of the polarization was reversed and not around (100) as suggested by infrared measurements. In addition, application of pulsed high intensity fields can be coupled to the pulsed neutron beam permitting investigation at fields much higher than can be obtained in steady state. An example is the investigation of $\alpha - \text{Fe}_2\text{O}_3$ with magnetic fields up to 120 kOe done at the Joint Institute of Nuclear Research in Dubna.¹⁷ Other studies that can be performed on the proposed facility include the study of relaxation phenomena in liquid crystals and biological substances by coupling pressure or temperature changes with the neutron pulses. In a similar fashion a beam of pulsed polarized neutrons can be used to investigate the relaxation phenomena in both single crystals and powders for protons oriented in a magnetic field with a selected r.f. pulse.¹⁸ A considerable effect can be observed using polarized neutrons because of the large difference in scattering length of oppositely oriented proton spins. Other studies include investigation of structural changes in materials under dynamic stress especially explosive stress, which can be examined by TOF due to the "simultaneous" observation of the entire diffraction pattern.

D. PROPOSED SINGLE CRYSTAL AND POWDER INSTRUMENTATION

There are unique problems related to single crystal diffractometry that limit the number of reflections that can be collected even with the most sophisticated of multidetector systems. Reference to the single crystal x-ray case is helpful in this regard. Consider the rotation camera where photographic film serves as a position sensitive detector. Reference to Fig. 1b shows that for the monochromatic beam case very few diffraction maxima occur for a crystal fixed in space. Although the number of diffraction maxima occurring at any one time is significantly increased with a polychromatic source, as Fig. 1a illustrates, only a small fraction of the total is observed. To obtain the other diffraction intensities, one must rotate, i.e., reorient, the crystal. Again, referring to Figs. 1a and 1b, in the monochromatic case, each reflection (within the k_{\max} sphere) would be observed once, while in the polychromatic case each reflection can be observed many times, with the reflections moving in toward the origin ($\theta = 0$) as the wavelength decreased. Alternately then, one could envision a narrow strip of film oriented parallel to the rotation axis and positioned in the backscattering region. A plane in the crystal would then diffract when d_{hkl} falls between d_{\min} and d_{\max} corresponding to λ_{\min} and λ_{\max} . Carrying this analogy over to the neutron diffractometer case, one could have an instrument with a single detector (the equivalent of only one small area element of film) capable of being appropriately positioned in space, as illustrated in Fig. 2, a linear detector approximately equivalent to the film strip mentioned above, or a complete multidetector facility.

A powder sample is composed in general of crystallites randomly oriented in space and therefore the orientation problems associated with single crystals disappear when powder samples are being used. In fact, in the polychromatic radiation case, only a few fixed detectors need be employed where each detector will record intensities of those reflections with d_{hkl} 's between d_{\min} and d_{\max} as selected by its θ value.

In practice, many detectors can be employed to advantage at different θ values to allow some overlap of ranges, and at the same θ value to improve counting statistics.

In the single crystal case, if something less than the very expensive complete multidetector assembly along with associated massive storage is to be used, the strategy employed for data collection becomes very important. In the monochromatic beam case, for example, a technique such as orienting layers of the reciprocal lattice parallel to the axis of position sensitive detectors and perpendicular to the rotation direction can very significantly reduce data collection times.¹⁹ One has considerably greater flexibility in the polychromatic case in this regard. However, it is still important to develop algorithms for the most efficient collection of high quality diffraction data to assure the shortest overall data collection times and to make optimal use of the assembly of detectors.

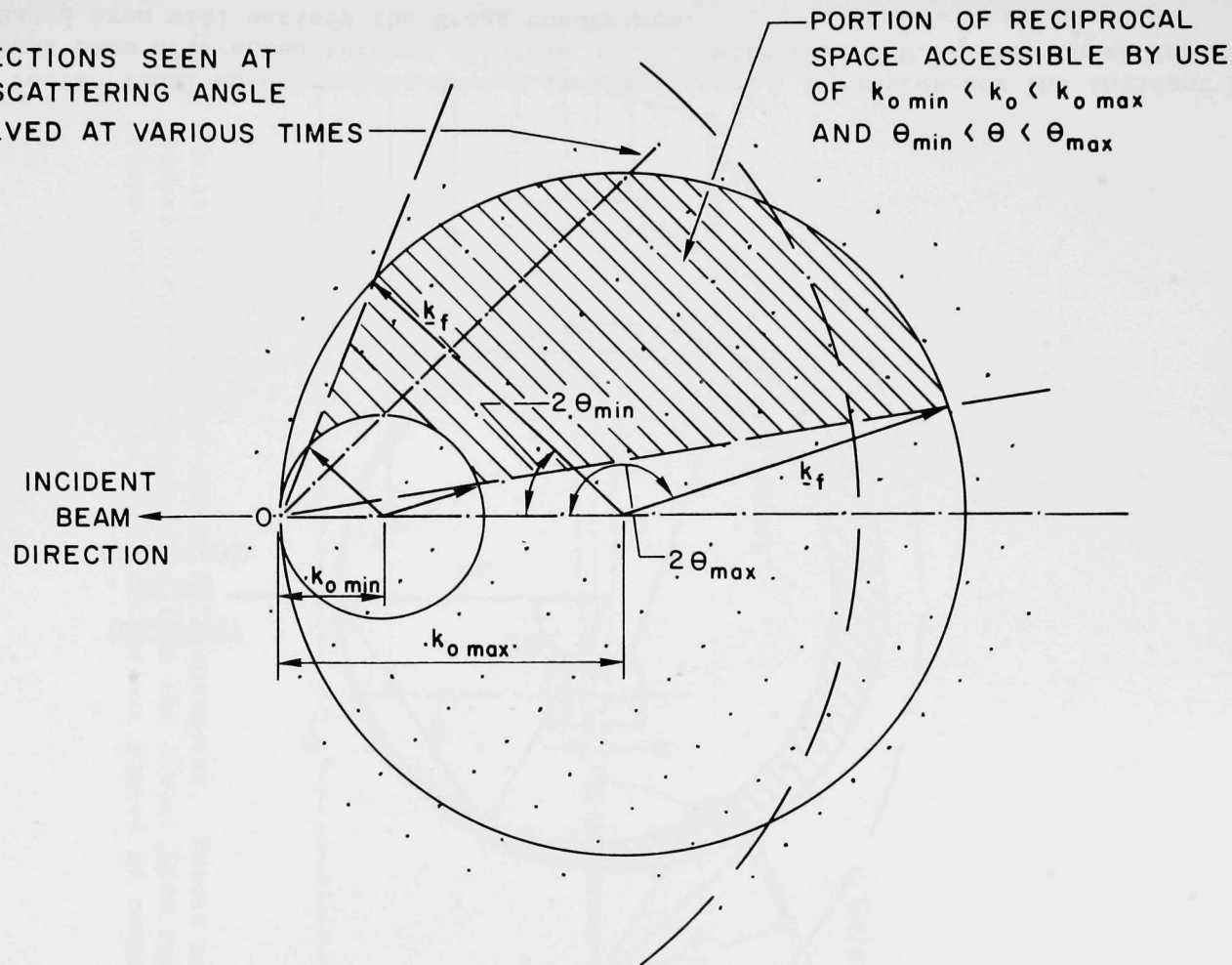


Fig. 1: Construction in reciprocal space to illustrate the use of white and monochromatic radiations in single crystal diffraction. In both figures the circle centered on the origin of reciprocal space represents the volume (or area in two dimensions) that must be measured to obtain a certain resolution in real space. In (a) the circles with radii $1/\lambda \min$ and $1/\lambda \max$ are drawn through the origin, and the direction of the radii represent the direction of the incident beam. All reciprocal lattice points within the shaded area will simultaneously diffract and may be samples by a large position-sensitive detector.

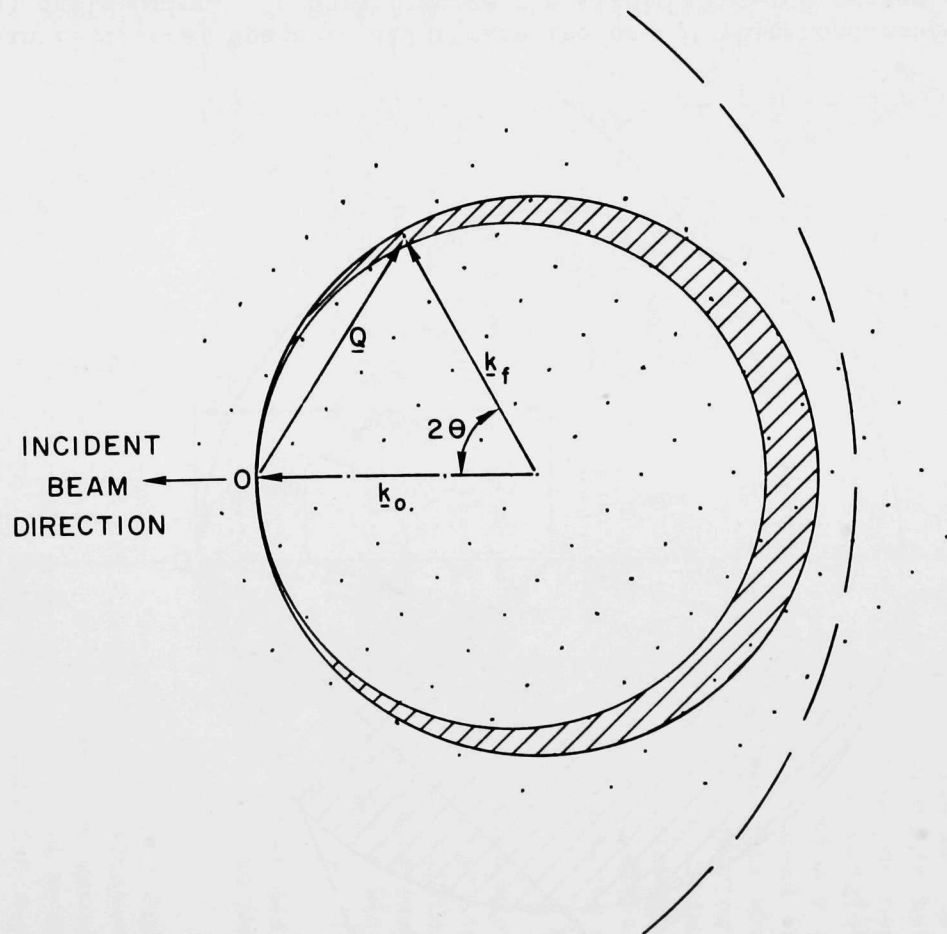


Fig. 1: (b) The conventional monochromatic system is illustrated. k_o represents the incident beam, k_f the final beam diffracted through an angle of 2θ . With $\Delta\lambda/\lambda \approx 0.02$ only those reflections in the shaded area will satisfy the Bragg conditions.

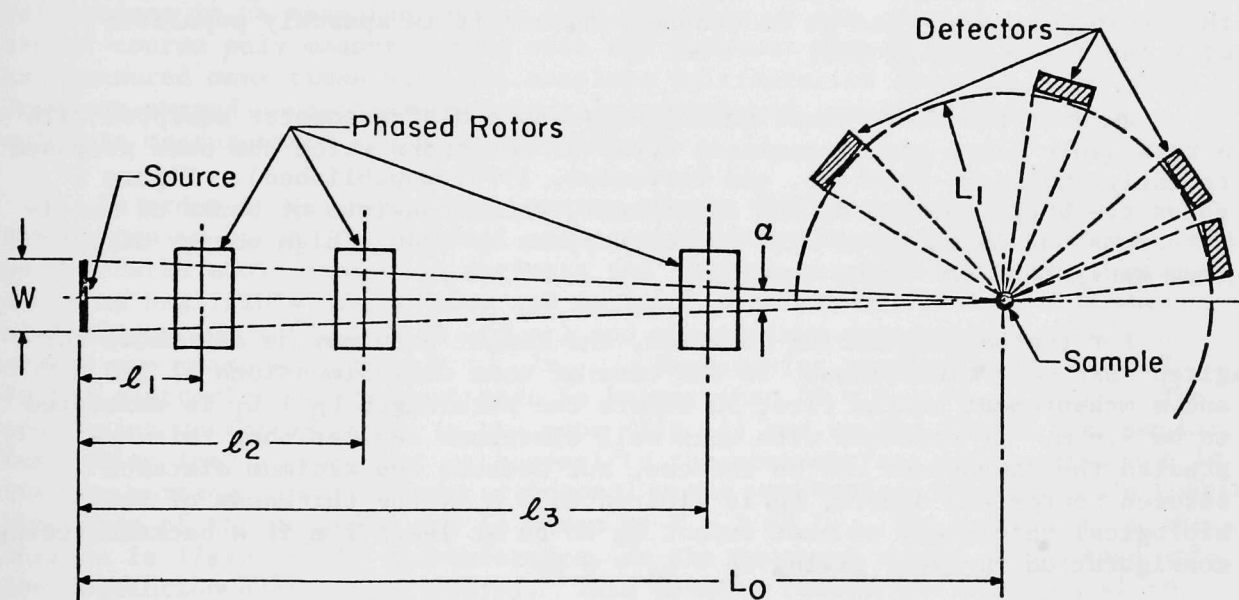


Fig. 2 Multipurpose time-of-flight diffractometer. Rotors are used to reduce high energy background from the direct beam and to minimize frame overlap. Banks of detectors are placed as convenient.

1. Single Crystal Diffractometers

Single crystal diffractometers can be roughly divided into two classes; namely, diffractometers for study of macromolecules, with unit cell dimensions above 40-50 Å, and diffractometers for study of small and medium size molecules.

An essential difference is in the number of reflections N to be recorded, which is approximately proportional to the unit cell volume. In addition, the physical environment of the sample during measurement should be variable for the small molecule instrument. Nonstandard values of pressure and temperature and external fields must be anticipated in this case whereas data collection on proteins commonly will take place near room temperature. A diffractometer designed for protein work may therefore not have optimal efficiency in other studies. We can however use approximately similar arguments concerning estimates of measuring time and machine geometry as the reciprocal space has to be scanned whether it is sparsely populated with lattice points or not.

We will therefore first briefly discuss a diffractometer equipped with a reasonably large two dimensional array of detectors which has been proposed recently (Meister, Peterson, and Carpenter, 1975, unpublished). Figure 2 shows the basic outline of the instrument, which consists of banks of multi-detectors and is equipped with three choppers to reduce high energy direct beam background and frame overlap.

For the backscattering position, the flight path may be estimated for given unit cell dimensions. In the case of unit cell dimensions of 100 Å and a measurement of the first 50 orders the pathlength $L_0 + L_1$ is estimated to be 9.6 m. If crystals with unit cell dimensions smaller than this are studied the dimensions can be reduced, but because the minimum distance between source and sample, L_0 , is limited to 5 m by the thickness of the biological shielding, we must expect L_0 to be at least 7 m if a backscattering configuration is used, giving $L_0 + L_1 = 9$ m.

At present it is difficult and expensive to setup a position sensitive detector with time resolution that will cover a large percentage of the space. However, limited size, 2-dimensional detectors are available of the order of 21×21 cm² with 900 detector elements, i.e., 0.7 cm resolution. For such a detector the measurement time can be estimated. Assuming cell dimensions as above and a crystal volume of 0.01 cm³ a measurement time of three days for one orientation of the crystal is expected, assuming 30% relative precision for the weakest reflections. A total of 5-10 settings of the crystal would be required. If the unit cell dimensions are approximately 20 Å, then measurement time approaches one hour per setting. In both cases a considerable advance over current technology is achieved.

As noted above, a complete multidetector assembly implies a very considerable expenditure for detector elements, their associated electronics and the massive storage device used. Only a percentage of the reciprocal lattice points are in position to diffract at any time and hence for a given position of the crystal, the great majority of detector elements will provide no useful information. It is appropriate then to ask if a simpler detector

arrangement could be employed without resulting in a significant decrease in information content.

Figure 3 shows that with a polychromatic source and a single detector, with time resolution capability, one obtains information concerning a line in reciprocal space, while with a complete two-dimensional multidetector assembly three-dimensional reciprocal space information is obtained. A linear position-sensitive detector of doughnut shape illustrated in Fig. 4 and placed in the forward scattering region appears to hold considerable promise. It can record reciprocal space information corresponding to those points falling on the surface of the cone indicated. In one rotation of the crystal, all reflections (k less than k_{\max}) would pass through this cone except for those very few reflections within a small angle, $\sim 10^\circ$, of the rotation axis. Provision for slight canting of the crystal would allow collection of all intensity data. Such a detector has far fewer elements than the complete multidetector assembly and yet allows essentially all reflections to be measured in one 180° rotation of the crystal. Reflections are of course only measured once with the doughnut-shaped detector while they are measured many times with the complete multidetector assembly. Two such doughnut-shaped detectors appropriately positioned could be utilized if multiple measurements would be desirable.

In order to estimate the geometry of the diffractometer for unit cell dimensions of about 20 \AA we will assume that the time resolution is sufficient as indicated above, and will estimate the spatial resolution for the detector assuming negligible sample size and no influence from the source. If the minimal spacing is τ_{\min} ($\tau = 1/d_{hkl}$) and the maximum spacing τ_{\max} , and n orders are measured with $n = \tau_{\max}/\tau_{\min}$. The angular resolution orthogonal to the scattering vector must then be better than $\Delta\theta = \tau_{\min}/\tau_{\max} = 1/n$. The scattering vector is lying in the cone (Fig. 4) and we may divide the angular resolution into a component orthogonal to the cone surface and a component in the cone surface. The component orthogonal to the cone surface determines the required thickness of the doughnut and is given by $T = 2 \Delta\theta L_1$. The component in the surface is $1/2\pi n$ of the circumference, so the doughnut should be divided into $2\pi n$ resolution elements or better. This gives a resolution of C_i equal to $2\pi L_1 (\sin 2\theta)/2\pi n = L_1 (\sin 2\theta)/n$.

Introducing a typical case with $L_1 = 1$ meter, $2\theta = 20^\circ$, unit cell dimensions of 20 \AA , $n = 30$, so that $\lambda_{\min} = 0.23 \text{ \AA}$ and $\lambda_{\max} = 6.9 \text{ \AA}$, and the real space resolution is 0.67 \AA , we get $T = 6.6 \text{ cm}$ and $C_i = 1.1 \text{ cm}$. These specifications for a detector seem reasonable in terms of present day technology.

An estimate of the time can be obtained using values from above mentioned 2-dimensional detector systems. One measurement will take about an hour. The rotation of the crystal about the vertical axis is determined by the detector thickness, i.e., $\Delta\theta$, so we get $57.3/30^\circ/\text{hour}$. 180 degrees rotation will then take 94 hours or approximately 4 days.

An estimate can be obtained too by comparison with a steady state high flux reactor. The time average flux for the pulsed source will be of the order of $10^9 \text{ n/cm}^2/\text{sec}$ for thermal neutrons compared to $10^7 \text{ n/cm}^2/\text{sec}$ for a monochromatic beam for a steady state source. On the other hand, a given reflection measured using the pulsed source will only use about 0.002 of the spectrum which gives a factor of 5 in disfavor of pulsed sources. Average

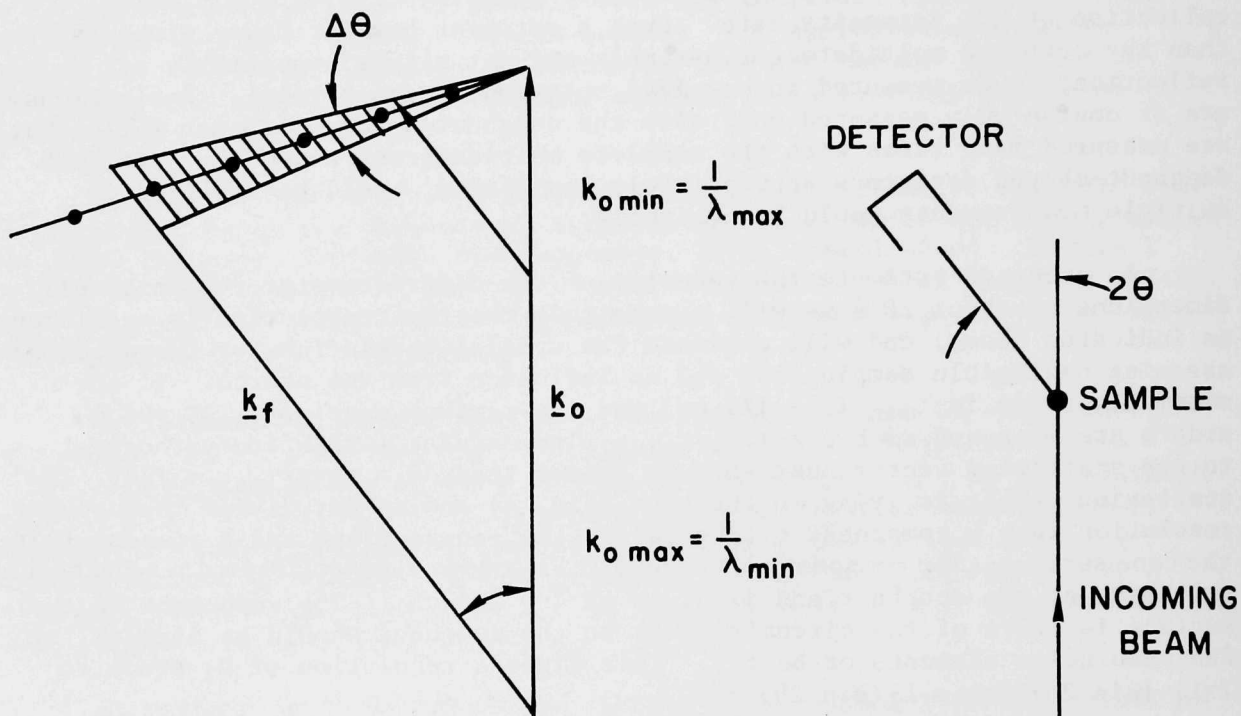


Fig. 3 Illustration of the part of the lattice in reciprocal space illuminated in the white beam technique using one detector covering an angular range of $2\Delta\theta$. k_o is the wavevector for the incoming beam, k_f is the wavevector for the diffracted beam. Lattice points within the shaded area originating from zero with radial limits defined by λ_{\min} and λ_{\max} can be observed, and if pulsed techniques are used, resolved. For forward scattering ($\theta = 0^\circ$) the central lattice lines illuminated will be orthogonal to the incoming beam.

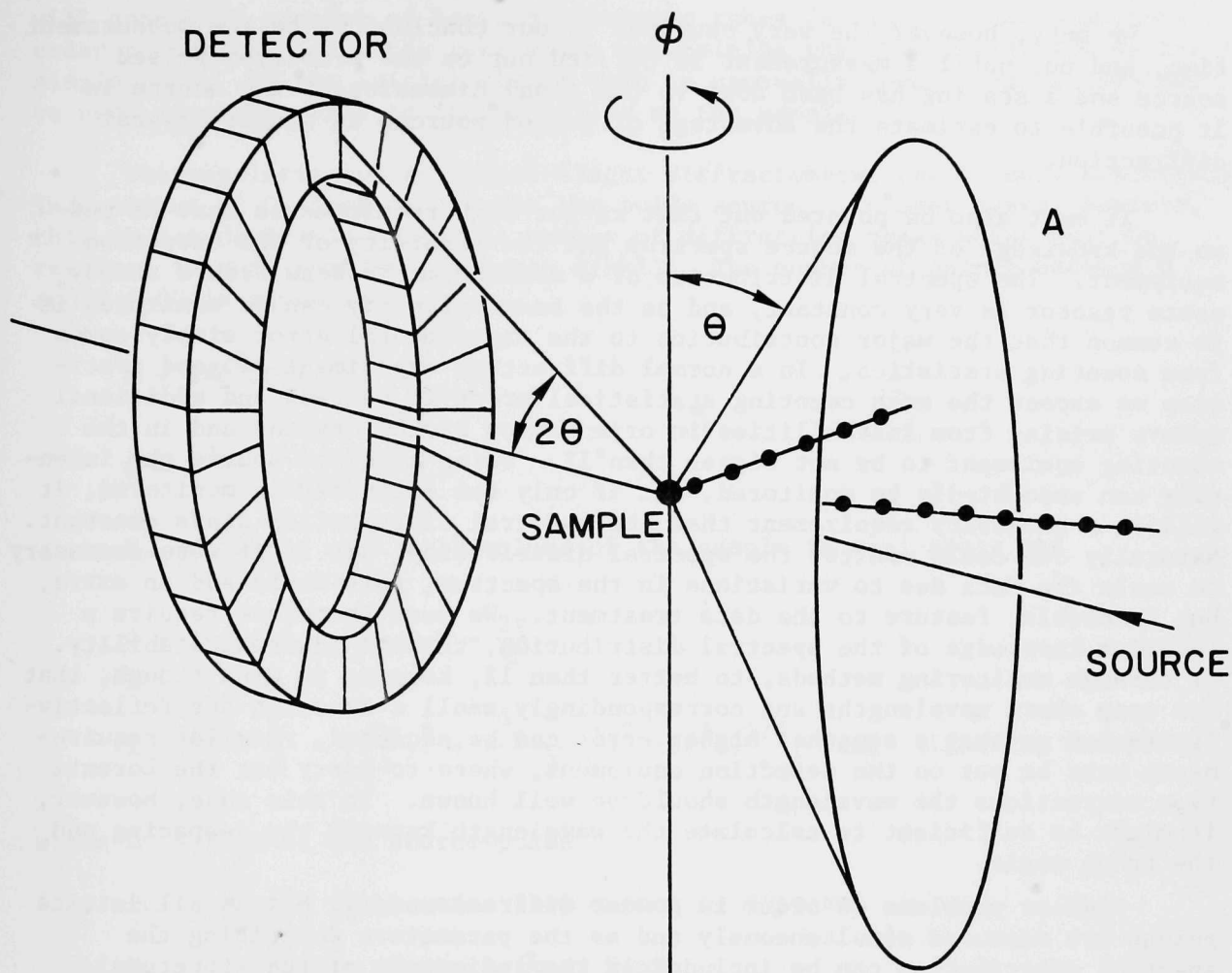


Fig. 4 Single crystal diffractometer. The doughnut-shaped detector can consist of a series of linear elements, c_i . The illuminated part of reciprocal space is indicated by the nearly flat cone having the apex at the origin of reciprocal space. Two central lines of lattice points are indicated, one (A) lying on the cone, giving rise to diffraction. The rotation axis ϕ is orthogonal to the incoming beam. One rotation of 180 degrees will bring all reflections into reflection position once, if the cone is flat. Elsewhere a few reflections near the ϕ axis will not be excited at any time.

measurement time to obtain a reflection on a steady state reactor to reasonable precision is 10 min so one orientation of the sample should be measured for $(5 \times 10/60)$ hour, giving $57.3/(30 \times 0.83)$ deg. per hour or 78 hours, i.e., 3 days for a full experiment.

We must, however, be very cautious in our conclusions on the measurement time, and not until a measurement is carried out on the prototype pulsed source and a scaling has been done to the final dimension of the source is it possible to estimate the advantage of pulsed sources in single crystal diffraction.

It must also be pointed out that rather high requirements must be put on the knowledge of the source spectrum and the stability of the detection equipment. The spectral distribution of a monochromatic beam from a steady state reactor is very constant, and as the beam intensity can be monitored it is common that the major contribution to the experimental error simply comes from counting statistics. In a normal diffraction experiment of good precision we expect the mean counting statistical error to be 2-3% and additional errors arising from instabilities in orientation of the crystal and in the counting equipment to be not bigger than 1%. Using a pulsed source the intensity can undoubtedly be monitored, but if only the intensity is monitored, it will be a necessary requirement that the spectral distribution stays constant. Naturally one could monitor the spectral distribution, but if it were necessary to scale the data due to variations in the spectrum, this would add an extra, but tractable, feature to the data treatment. We must therefore require a relative knowledge of the spectral distribution, through inherent stability, or through monitoring methods, to better than 1%, keeping in mind though, that for very short wavelengths and correspondingly small d-spacings the reflectivity is low so that a somewhat higher error can be accepted. Similar requirements must be put on the detection equipment, where to carry out the Lorentz type corrections the wavelength should be well known. In this case, however, it might be sufficient to calculate the wavelength knowing the d-spacing and the Bragg angle.

Similar problems do occur in powder diffractometry, but as all lattice points are measured simultaneously and as the parameters describing the spectral distribution can be included in the refinement of the structural parameters only the time averaged distribution is of interest, thereby possibly reducing our requirements.

2. Powder Diffractometers

A pulsed source with a repetition rate in the range of 30-60 hz is ideally suited for high resolution time-of-flight powder diffractometry. In such an instrument, the total path length would be relatively long (10-100 m) in order to minimize resolution contributions from time uncertainties. This long path length requires that the repetition rate of the source pulses be rather low (10-100 hz) in order to allow a suitable region of Q space to be scanned at a single scattering angle. The resolution of the instrument is primarily determined by the pulse width. Powder diffractometers can, therefore, readily be designed to take full advantage of the high peak neutron flux available at a pulsed source while also providing the degree of resolution required for the proposed experiments.

A typical time-of-flight powder diffractometer is shown in Fig. 2.

L_0 is the length of the initial flight path from source to sample and L_1 is the distance from sample to detector. Rotating collimators may be desired to eliminate frame overlap in some instruments. Detector arrangements will generally consist of banks of detector tubes in time focused positions in order to increase counting rate while maintaining the same resolution as for a single tube. If the initial flight path is unusually long, a guide tube may be used advantageously to increase flux at the sample.

The resolution of a time-of-flight diffractometer can be made essentially independent of Q since $\Delta t \sim \lambda$ for the pulse source. In most cases, however, this is not desirable since the number of diffraction peaks which must be resolved is much smaller for large $d(hkl)$. The number of points between Q and $Q + \Delta Q$ is given by

$$N = 4\pi Q^2 \Delta Q / [(2\pi)^3 / V_{\text{cell}}]$$

$$N = Q^2 \Delta Q V_{\text{cell}} / 2\pi^2 ,$$

where V_{cell} is the unit cell volume of the sample in real space and

$$Q = 2k \sin\theta = \frac{4\pi}{\lambda} \sin\theta = \frac{2\pi}{d} .$$

$$R = \frac{\Delta d}{d} = \frac{\Delta Q}{Q} = [(\frac{\Delta t}{t})^2 + (\frac{\alpha_0 + \alpha_1}{2})^2 \cot^2\theta]^{1/2}, \text{ and (ignoring focussing)}$$

where Δt = FWHM of the source pulse

t = neutron flight time from source to detector

α_0 = divergence of the primary beam

α_1 = divergence of the secondary beam.

In order to resolve the points we require $N = 1$ or,

$$8k^3 \sin^3\theta [(\frac{\Delta t}{t})^2 + (\frac{\alpha_0 + \alpha_1}{2})^2 \cot^2\theta]^{1/2} = \frac{2\pi^2}{V_{\text{cell}}}$$

The Intensity (neutrons/sec) of a time-of-flight diffractometer is given by:

$$I = A_{\text{source}} * \phi_{\text{source}} * \frac{A_{\text{sample}}}{4\pi L_0^2} * \text{GTG} * \frac{A_{\text{det}}}{4\pi L_1^2} * \text{SR} , \quad (1a)$$

where the time average source flux is

$$\phi_{\text{source}} = 2.7 \times 10^{13} \text{ n/cm}^2 \text{ sec (for IPNS moderator C)}$$

$$A_{\text{source}} = 100 \text{ cm}^2 \text{ if the full source is viewed}$$

GTG = guide tube gain =

$$(1 + (\frac{\theta_c}{\phi_{\text{max}}})^2)(1 + (\frac{\theta_c}{\psi_{\text{max}}})^2); \quad 0 \leq \theta_c \leq \phi_{\text{max}}$$

$$(\frac{2\theta_c}{\phi_{\text{max}}})(1 + (\frac{\theta_c}{\psi_{\text{max}}})^2); \quad \phi_{\text{max}} \leq \theta_c \leq \psi_{\text{max}}$$

$$\frac{4\theta_c^2}{\phi_{\text{max}}\psi_{\text{max}}}; \quad \psi_{\text{max}} \leq \theta_c \quad (1b)$$

where $\phi_{\text{max}} = W/L_0$, $\psi_{\text{max}} = H/L_0$ are the horizontal and vertical angular divergences of the guide, H and W the width and height of the guide aperture, and it is assumed that $H \geq W$. The critical angle is $\theta_c = \alpha_c \lambda$, where $\alpha_c = 1.75 \times 10^{-3} \text{ rad/\AA}$ for a nickel coated guide.

The last factor in Eq. (1), SR , is the sample reflectivity which depends on sample thickness and cross sections, and as used here is an average over the spectrum.

Using these ideas, performance estimates have been made for three diffractometers.

a. High Resolution Diffractometer (HRD)

This instrument is designed to study polycrystalline samples with as many as approximately 10,000 reflections at lattice spacings greater than 0.8 \AA . This will make this instrument capable of studying samples that have heretofore only been amenable to single crystal studies. It is expected that the data collection time will be much shorter than that required for a single crystal sample and the studies will not be restricted to samples for which single crystals are available.

Design Goals

This diffractometer should provide a resolution of $\Delta d/d \sim 0.1\%$ for a minimum d -spacing of $d_{\text{min}} = 0.8 \text{ \AA}$ with $Q_{\text{max}} = 2\pi/d_{\text{min}} = 7.85 \text{ \AA}^{-1}$. It should also be capable of resolving lines for larger lattice spacings of the same crystal and should extend out to at least $d_{\text{max}} \sim 6 \text{ \AA}$.

The path lengths required are:

$$L_0 = 98.5 \text{ m}$$

$$L_1 = 1.5 \text{ m}$$

Time focused detector banks are placed at several different angles as shown in Table I.

Table I

θ_B	d_{\min}	d_{\max}	Q_{\min}	Q_{\max}	$\Delta d/d$
88°	0.8	1.466	4.286	7.854	0.001
33.2°	1.460	2.674	2.350	4.304	0.0033
17.7°	2.627	4.819	1.304	2.392	0.0108
9.73°	4.735	8.668	0.725	1.327	0.3500
5.39°	8.516	15.59	0.403	0.738	0.1133
3°	15.32	27.99	0.224	0.410	

$\lambda_{\min} = 1.6 \text{ \AA}$; $\lambda_{\max} = 2.93 \text{ \AA}$, with a repetition rate of 30 Hz.

The data collection rate for this instrument has been estimated by comparison with the powder diffractometer at ZING P2. Assuming a sample of volume 1 cm^3 and a detector area (at a single scattering angle) of 0.013 steradians, the rate for this high resolution diffractometer would be about 2000 times that achieved for the ZING P2 diffractometer.

b. Small Sample High Intensity Diffractometer (HID)

This instrument is proposed as having medium resolution ($\Delta Q/Q \sim 1.2\%$) and data collection rates high enough to allow the study of samples as small as 1-10 milligrams. The initial and final flight paths are: $L_0 = 9 \text{ m}$, $L_1 = 1 \text{ m}$. By placing detector banks at scattering angles of $2\theta = 90^\circ$ and $2\theta = 30^\circ$, it would be possible to measure d spacings from 0.35 to 9.67 \AA , utilizing neutrons of wavelengths between 0.5 and 5 \AA . To achieve the desired counting rate approximately 100 detectors ($2.54 \text{ cm diam} \times 25 \text{ cm long}$) would be required at each scattering angle. A comparison with counting rates at ZING P2 yields the conclusion that for a 1 mg sample the required counting time would be about 1 day.

c. High Pressure Diffractometer

The design of a high pressure diffractometer is in many respects analogous to the design of a high intensity, small sample instrument. In the high pressure case, the sample volume subtended by the source and the detector is severely limited by the small window size available in the pressure cell. Time-of-flight techniques are ideally suited for diffraction at high pressure because realiable window dimensions are not sufficient for angular scan techniques. The high pressure diffractometer proposed for IPNS is intended for use with a cubic press capable of pressure to approximately 100 kilobars and temperature above 1000°C . This geometry requires data collection at a single scattering angle of $2\theta = 90^\circ$. Neutrons will enter and leave the high pressure region through the gasket areas between the press anvils. The window dimensions will be about 0.1 cm wide by 1.0 cm high, giving an effective sample volume of 0.01 cm^3 . A comparison with the small sample, high intensity diffractometer shows that it is feasible to achieve resolution below $\Delta Q/Q = 0.5\%$ for the high pressure instrument with data collection times of the order of one day.

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IV. REPORT OF THE PANEL ON CHEMICAL STRUCTURES OF DISORDERED SOLIDS AND
INHOMOGENEOUS SYSTEMS AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. MATERIALS PROBLEMS

1. Introduction

In the context of this report, materials are taken to include polymers, alloys, ceramics and glasses, including amorphous semiconductors. The kind of problem we are concerned with keys on the relationship of structure and properties which is the central dogma of materials science and engineering. This relationship needs to be understood as completely and quickly as possible in view of the materials limited problems in such areas as the development of new energy sources. A new intense pulsed-neutron source can provide information in a number of ways that cannot at present be supplied with any other materials resource. First and foremost among these is the study of distortions or atomic displacements of solute atoms and solute clusters that occur in dilute or concentrated solid solutions, due to interstitials or due to radiation damage. Apart from the intense Huang scattering near Bragg peaks which gives information on the size and shape of the clusters, information on the near-neighbor environment of the defects is obtained by investigating the elastic diffuse scattering between the Bragg peaks.

2. The Cross-Section for Elastic Diffuse Scattering

The cross section for elastic diffuse scattering for the case of not too strongly distorting defects may be written as: (see e.g., Ref. 1, Eq. (9))

$$\left. \frac{d\sigma}{d\Omega} \right|_{\text{elastic}} = N |\Delta\tilde{C}(\vec{k}) [f_D(\vec{k}) + \bar{f}(\vec{k}) i\vec{k} \cdot \vec{S}(\vec{k})]|^2. \quad (1)$$

The concentration fluctuations are contained in $\Delta\tilde{C}(\vec{k})$; $f_D(\vec{k})$ is the scattering amplitude of the defect and contains information on the defect position, and $\vec{S}(\vec{k})$ is the Fourier transformation of the distortion field, $\bar{f}(\vec{k})$ is the average scattering amplitude. To illustrate the difficulties encountered in trying to analyze the data measured with x-rays, we use the following formulation for a cubic metallic alloy with one atom per lattice point² in which thermal displacements are also taken into account, because they cannot be discriminated against in a measurement with x-rays; this information also includes mean-square static displacements more completely than Eq. (1).

$$\begin{aligned} I_D(h_1, h_2, h_3)/N &= I_{SRO}(h_1, h_2, h_3) \\ &+ h_1 Q_x(h_1, h_2, h_3) + h_2 Q_x(h_2, h_3, h_1) + h_3 Q_x(h_3, h_1, h_2) \\ &+ h_1^2 R_x(h_1, h_2, h_3) + h_2^2 R_x(h_2, h_3, h_1) + h_3^2 R_x(h_3, h_1, h_2) \\ &+ h_1 h_2 S_{xy}(h_1, h_2, h_3) + h_2 h_3 S_{xy}(h_2, h_3, h_1) \\ &+ h_3 h_1 S_{xy}(h_3, h_1, h_2), \end{aligned} \quad (2a)$$

where h_i are the coordinates in reciprocal space and N is the number of atoms in the sample. With X_μ as the atomic fraction of the μ th component:

$$I_D(h_1, h_2, h_3) = \frac{[I_{\text{Total}}(h_1, h_2, h_3) - I_{\text{Bragg}}(h_1, h_2, h_3)]}{X_A X_B (f_A - f_B)^2} \quad (2b)$$

$$I_{\text{SRO}}(h_1, h_2, h_3) = \sum_l \sum_m \sum_n \alpha_{lmn} \cos \pi l h_1 \cos \pi m h_2 \cos \pi n h_3, \quad (2c)$$

$$Q_x(h_1, h_2, h_3) = \sum_l \sum_m \sum_n \gamma_{lmn}^x \sin \pi l h_1 \cos \pi m h_2 \cos \pi n h_3, \quad (2d)$$

$$R_x(h_1, h_2, h_3) = \sum_l \sum_m \sum_n \delta_{lmn}^x \cos \pi l h_1 \cos \pi m h_2 \cos \pi n h_3, \quad (2e)$$

$$S_{xy}(h_1, h_2, h_3) = \sum_l \sum_m \sum_n \epsilon_{lmn}^{xy} \sin \pi l h_1 \sin \pi m h_2 \cos \pi n h_3. \quad (2f)$$

The "Fourier coefficients" of the individual diffuse intensity components, Q_x , R_x and S_{xy} , are related to the moments of the displacements in the lattice (static plus thermal) by the expressions:

$$\gamma_{lmn}^x = 2\pi [F_{AA} \langle x_{lmn}^{AA} \rangle + F_{AB} \langle x_{lmn}^{AB} \rangle + F_{BB} \langle x_{lmn}^{BB} \rangle], \quad (2g)$$

$$\delta_{lmn}^x = 2\pi^2 [F_{AA} \langle (x_{lmn}^{AA})^2 \rangle + F_{AB} \langle (x_{lmn}^{AB})^2 \rangle + F_{BB} \langle (x_{lmn}^{BB})^2 \rangle], \quad (2h)$$

$$\epsilon_{lmn}^{xy} = 4\pi^2 [F_{AA} \langle x_{lmn}^{AA} y_{lmn}^{AA} \rangle + F_{AB} \langle x_{lmn}^{AB} y_{lmn}^{AB} \rangle + F_{BB} \langle x_{lmn}^{BB} y_{lmn}^{BB} \rangle], \quad (2i)$$

where

$$F_{AA} = \frac{f_A^2}{(f_A - f_B)^2} (X_A/X_B + \alpha_{lmn}), \quad (2j)$$

$$F_{AB} = \frac{2f_A f_B}{(f_A - f_B)^2} (1 - \alpha_{lmn}) , \quad (2k)$$

$$F_{BB} = \frac{f_B^2}{(f_A - f_B)^2} (X_B/X_A + \alpha_{lmn}) . \quad (2l)$$

The quantity $\langle x_{lmn}^{\mu\nu} \rangle$ denotes the average displacement along the [100] axis between μ - ν atom pairs separated by the vector r_{lmn} , with similar definitions for the quantities $\langle (x_{lmn}^{\mu\nu})^2 \rangle$ and $\langle x_{lmn}^{\mu\nu} y_{lmn}^{\mu\nu} \rangle$.

Only terms in the average x components of displacement appear, the other components being related to this by cubic symmetry, e.g., $\langle x_{lmn}^{\mu\nu} \rangle = \langle y_{lmn}^{\mu\nu} \rangle$.

The Fourier coefficient α_{lmn} is the normal Warren short-range order parameter relating $P_{lmn}^{A/B}$, the conditional probability of finding a B atom at the end of the vector r_{lmn} , if there is an A atom at the origin of the vector:

$$\alpha_{lmn} = 1 - \frac{P_{lmn}^{A/B}}{X_B} . \quad (2m)$$

3. Short Range Order, Distortions, Clustering

The first term in Eq. (1a) I_{SRO} peaks near Bragg reflections for clustering, or between them for local order. The terms in the expansion for Q are modulations in intensity which change sign at the nodes of the reciprocal lattice. The displacement terms R and S peak strongest near the Bragg reflections--the Huang scattering referred to above plus first order thermal diffuse scattering (TDS). If it could be assumed that the coefficients in Eqs. (2b)-(2f) were independent of h then the terms can be separated because of their different symmetries in reciprocal space (see Ref. 2). Unfortunately, with x-rays, the scattering factor ratios included in these coefficients (Eqs. (2g)-(2l)) vary considerably in reciprocal space and the separation is only approximate at best.* This is not a problem with neutrons. Furthermore, there is an inelastic scattering process that can contribute significant intensities at low or moderator scattering angles as well as first order TDS. This is the interference between one and two-phonon processes.⁴⁻⁶ It is a modulation similar to the Q terms, but, depending on the system may have the same or opposite in sign as Q . With energy analysis of the scattering neutrons, this problem can be eliminated.

Knowledge of the distortions is vital to our understanding of the mechanical, electrical and magnetic behavior of materials--and we just cannot do a proper job with x-rays. With an intense neutron source a proper separation can finally be attempted. All of these scattering contributions are

* For a detailed discussion, see Ref. 3.

weak, compared to contributions due to background and phonon scattering as demonstrated in Fig. 1. In Fig. 1, E refers to the small elastic scattering of interest. Therefore, an intense neutron source is needed, especially for dilute solutions.

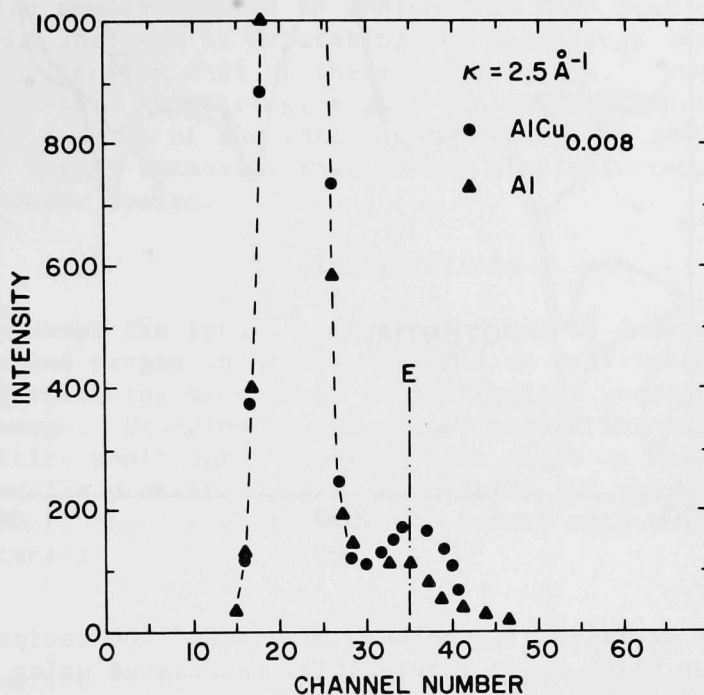


Fig. 1: TOF-spectra taken with AlCu_{0.008} and pure Al at T = 800 K. The peak around E is elastic scattering; inelastic scattering shows up around channel 20 (Ref. 7). Intensity vs. channel.

In addition, previous studies carried out primarily with x-rays have for the most part concentrated on model systems. As a result, little is known about local atomic arrangements of ferrous alloys because the x-ray scattering factors of the components are quite similar, since the elements involved are neighbors in the periodic table. The neutron scattering factors for these elements are usually not similar and furthermore, the differences in neutron scattering factors can be enhanced or reduced by isotopic enrichment. In many materials, however, it is difficult to make specimens large enough to study with conventional neutron sources and an exceptionally intense source is required.

4. Interatomic Potentials

Another important area is that of the determination of interatomic potentials in alloys and ceramics. As the scattering pattern is related to the transform of the potential, the diffuse scattering may be examined to test and to measure such potentials. There is increased interest in such measurements for the development of alloy theory. As an example, we show scattering curves for self-interstitials in tungsten calculated for two different potentials (see Fig. 2). It is evident that, in order to measure such differences, good accuracy in the scattering data is required. This again amounts to a need for a high intensity spectrometer. Not only is the

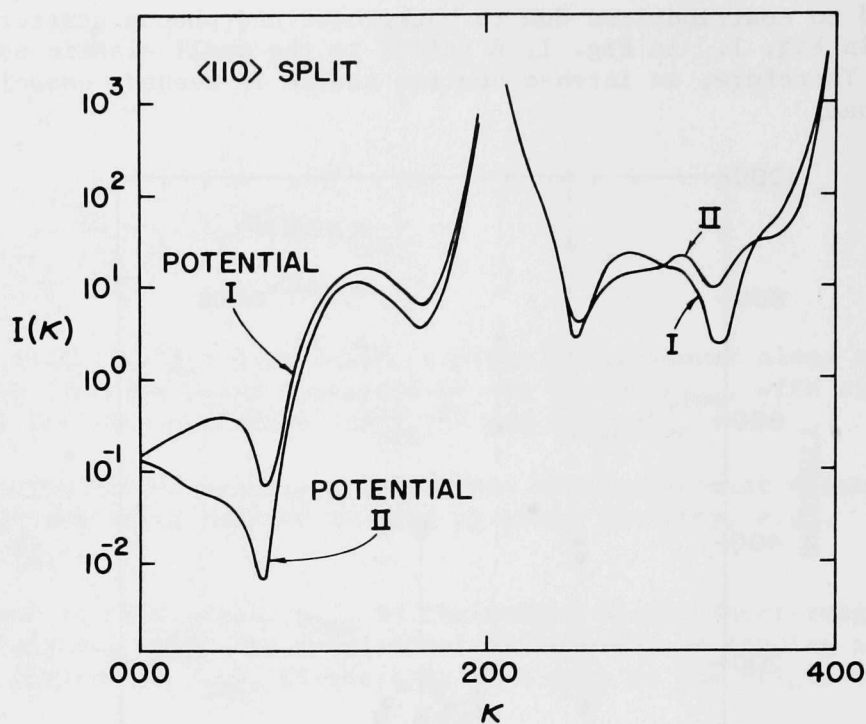


Fig. 2: Intensity profiles $I(\vec{q})$ between origin and 400 reciprocal lattice vector for $\langle 110 \rangle$ split interstitial calculated using two different interatomic potentials (Ref. 8).

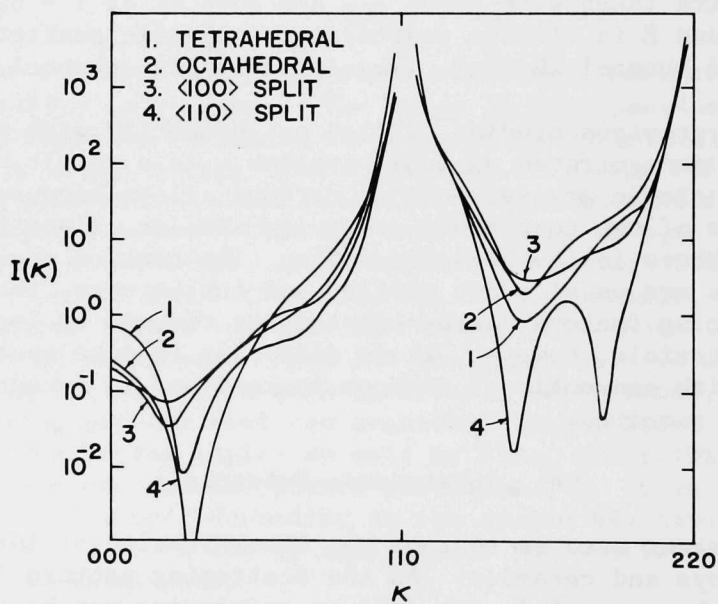


Fig. 3: Intensity profiles $I(\vec{q})$ between origin and 220 reciprocal lattice vector for tetrahedral, octahedral, $\langle 100 \rangle$ split and $\langle 110 \rangle$ split configurations in tungsten (Ref. 8).

self-potential important, but also the potential between different constituents of an alloy.

For binary alloys such measurements have been undertaken at steady state reactors utilizing monochromators in conjunction with beam chopping techniques and time-of-flight analysis in separating the relatively large inelastic scattering components from that of the elastic regime.¹ Due to the very low cross sections for this scattering, a general application of these methods in investigating the details of the interatomic potentials seems to require a pulsed source of higher intensity than is now available coupled with an advanced spectrometer design.

5. Interstitials

Information about the location (type of site) of interstitials such as carbon, nitrogen and oxygen in metals, as well as self interstitials, is important in understanding mechanisms for diffusion, phase transformations and radiation damage. Examples of calculated scattering cross sections for various interstitial positions in tungsten are shown in Fig. 3. Because these defects are generally present in small quantities (of the order of a few tenths of a percent) these are difficult to detect with conventional x-ray or neutron scattering.

6. Kinetics

Much information is needed on the kinetics of local atomic arrangement--ordering, clustering, transformations, that could be supplied by studies of diffuse scattering as a function of time from a pulsed source.

7. Precipitates

Many properties of defect clusters and precipitates can be conveniently studied by x-rays through high resolution Huang diffuse scattering measurements near Bragg reflections and by small-angle scattering. However, x-ray measurements carried out between the Bragg reflections to investigate the atomic details within such defects in low concentrations mainly confined to very low temperatures due to the influence of thermal diffuse scattering. The details of precipitate nucleation and growth at elevated temperatures seem therefore to require methods where the elastic components can be recovered. This now becomes a possibility for the first time with the pulsed neutron source and the special time-of-flight spectrometer described in the second part of the paper.

8. Polymers, Gels, Rubbers

For polymers, gels, and rubbers, one can study the spatial correlations of adjacent atoms. Correlation functions in this range are, of course, available from neutron scattering data in momentum transfer ranges $> 0.1 \text{ \AA}^{-1}$. In this range, polymers, gels and rubbers do not differ in any qualitative way from other disordered solids. However, for longer distances corresponding to momentum transfers of 1×10^{-4} to $1 \times 10^{-1} \text{ \AA}^{-1}$ (the small-angle scattering region), there exist spatial correlations to polymeric systems. Polymers can be made having molecular weights of several million. Thus, two atoms separated even by as much as several thousand \AA could be correlated by being on the same

molecule. Two quantities of interest in this regard are the radius of gyration of a single molecule R_g and the persistence length P . In many solid polymers and in polymer solutions, R_g for a single chain is a simple function of the molecular weight being proportional to the molecular weight to a power β . The value of β depends upon the background material, having different values for dilute solutions, concentrated solutions and the bulk polymer. The persistence length is the average distance of a single polymer in a specific direction. This quantity is essentially independent of the molecular weight, and is usually smaller than the radius of gyration.

In order to measure either of these quantities, the molecule of interest must have a contrast with respect to the background material for neutron scattering. For dilute solutions, a contrast is present since the polymer and solvent generally have different chemical compositions. For these, the only differences between small-angle neutron scattering and small-angle x-ray scattering or light scattering are the contrasts and the available ranges of momentum transfer. The first small-angle neutron scattering experiments on polymer systems were done on dilute polymer solutions of polystyrene in carbon disulfide. In these cases the contrast for neutrons, was much greater than that for x-rays. The radii of gyration would not have been obtained for these solutions by small angle x-ray scattering because of the low contrast. The momentum transfers for light scattering experiments are too low to measure the radius of gyration of these solutions.

One can observe the single-chain spatial conformation in concentrated solutions or in bulk, by "tagging" a fraction of the molecules (deuteration) in a protonated host (or vice versa). Success depends upon hydrogen isotopic substitution not altering the chemical structure and the contrast factor between "tagged" and untagged chains being very strong.* Of course, one assumes here that the "tagged" molecule does not distort the surroundings. Neutron data has recently shown that for pure amorphous bulk polymer, the chain conformation fulfills the Flory prediction that each molecule sees its surrounding bulk as though it were in a θ solvent in which $R_g \sim M_w^{1/2}$.

Neutron experiments are now being applied to more complex polymeric systems with considerable success. In particular, there are current measurements of the radius of gyration for bulk crystalline polymers, orientation effects in drawn bulk polymers and ordering in solutions of copolymers.

The SANS (small-angle neutron scattering) experimental technique is a simple one in principle but demanding in intensity and in the momentum transfer range available, the latter from 10^{-4} to perhaps 10^{-1} \AA^{-1} . The best method requires a long wavelength ($> 4 \text{ \AA}$) high intensity collimated source, monochromated to the order of 10% only, impinging on the target. The scattered beam is most effectively observed by a two-dimensional detector array.

There may be some real advantages to performing SANS experiments with IPNS. For equal time-averaged source fluxes at the source plane (IPNS versus steady state reactor) a much higher counting rate can be achieved by using a

* The relevant cross section is given by Eq. (1) with \tilde{C} being the concentration of deuterated polymer.

broad region of the source spectrum (assuming that spectrum is well known and stable). There may also be a real advantage of broadening the useful $|\vec{k}|$ interval for a fixed square detector array by allowing the velocity and the scattering angle to vary.

Summary of Problems

1. Solid solution (statics and kinetics)
 - a. Distortion (include radiation induced)
 - b. Interatomic potentials
 - c. Clustering, SRO
 - d. Precipitation
2. Phase transformation (metals, ceramics, polymers, include glasses)
3. Polymers
 - a. Molecular configurations in dilute solutions
 - b. Tagged molecules-molecular configurations in concentrated solutions and in bulk.

Problem	Comment
Interstitials, dilute solution, defect interactions	Elastic scattering between Bragg peaks
Concentrated alloys, ordering	Extended region in κ out to at least 8 \AA^{-1}
Kinetics of precipitation and phase transformation	Short time measurements at low and intermediate κ -values $2-0.001 \text{ \AA}^{-1}$
Polymers	Very small angle elastic scattering at κ -values 10^{-1} to 10^{-4} \AA^{-1}

B. MACHINE DESIGNS

We present two instruments designed especially to examine the weak diffuse scattering between Bragg peaks and at small angles; these have been conceived in such a way as to make them of maximum use to a broad community. The committee wishes to especially thank one of its members, Dr. G. Bauer. His extensive background in neutron scattering and associated equipment design and his fertile ideas in this field provided our primary impetus.

1. Medium Resolution High Intensity Spectrometer for Diffuse Scattering

a. General Experimental Considerations

From experiments done so far on spectrometers especially designed for elastic diffuse scattering¹ some serious limitations of the existing instruments become evident, which emerge from the intensity available as well as from the limited range in momentum space that can be investigated.

Due to the obvious need for energy analysis of the scattered neutrons, the spectrometers are operated in a TOF-mode because it was felt that this was the best way to optimize the detected intensity by matching the resolution in energy and momentum transfer to the expected smooth variation of the elastic scattering between the Bragg peaks. It turns out, however, that a somewhat better resolution (of the order of 1-2% in κ) is desirable in many cases to minimize the influence of Bragg peaks on the one hand and to be able to investigate the width of fairly sharp structures, which occasionally show up as, for example, at the [110]-position in Fig. 4.

With a steady-state neutron source any improvement in resolution will be at least partly at the expense of the duty cycle of the chopper. This is not true for a pulsed source as its pulse length is short enough.

We think that the way to make optimum use of the pulsed source in the design of a spectrometer for elastic diffuse scattering is to operate the instrument in a TOF mode and monochromatize the scattered neutrons rather than the incident beam. The concept of such a spectrometer is developed in the following sections.

b. Basic Design Idea

We first give a brief description of the mode of operation of the proposed instrument by referring to Fig. 5.

A short pulse emitted from the source P will spread out while traveling along the distance L in such a way that different energies arrive at the sample S at different times. With the analyzer A set up such that it only transmits neutrons of one wavelength to the sample at any given time, only such neutrons will be recorded which have one outgoing wavelength λ_1 . The time of flight between sample and detector therefore is equal for all neutrons detected. It is therefore possible from the TOF spectrum recorded to assign a wavelength to each neutron, which it had before the scattering event and thus get the energy transfer in the scattering event. In particular, those neutrons can be extracted which underwent elastic scattering.

c. Extension to Analyzer Systems

To make optimum use of scattered neutrons, two innovations are needed.

- i) The most obvious extension is to use many analyzer-detector arrangements at the same time, set up a different scattering angles around the sample. Such an arrangement is shown in Fig. 6.

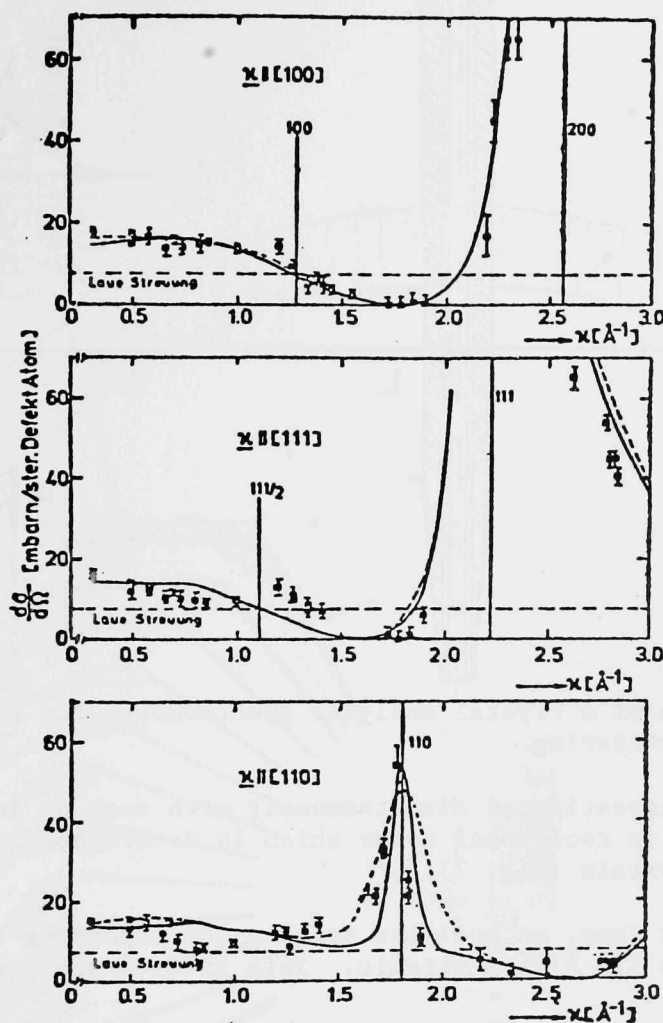


Fig. 4. Elastic diffuse scattering cross section for Bi in Pb for the three directions of lattice symmetry. The sharp peak at $\kappa = \tau^{220}/2$ has been attributed to a non-centro-symmetric force distribution around the Bi-atom (Ref. 9).

Its small width can presently only be understood by assuming orientation correlation in the displacements around the Bi-atoms (Ref. 10).

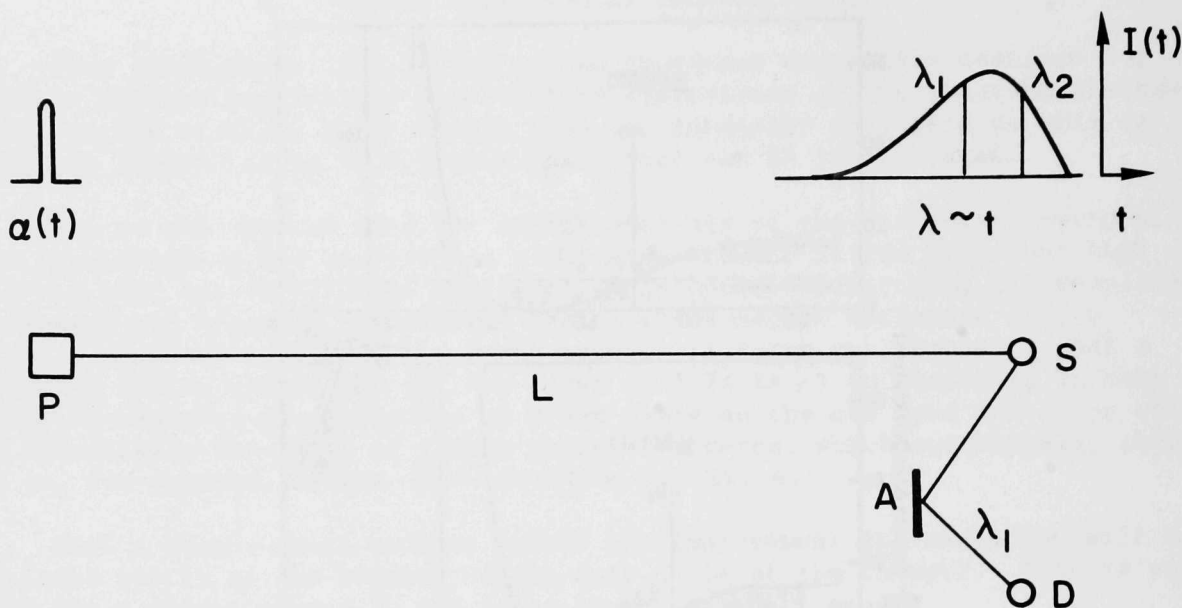


Fig. 5: Principles of a crystal analyzer spectrometer for elastic diffuse scattering.

The κ -values investigated simultaneously with such an instrument lie on an Ewald circle in reciprocal space which is determined by the setting of the analyzer crystals (Fig. 7).

In the present case, an analyzer setting corresponding to a transmitted wavelength of about 1.5 Å is desirable. This gives a scattering angle of about 30°.

ii) To make optimum use of the incident spectrum, a second extension should be considered. Inserting more than one crystal in each scattered beam allows one to adjust each of the crystals to a different energy and thus a larger fraction of the total beam intensity can be utilized. The arrangement for one arm is shown schematically in Fig. 8a; Fig. 8b gives the corresponding diagram in momentum space, for elastic scattering.

In this case the κ values investigated simultaneously are parallel to each other but pertain to different Ewald spheres.

It is likely that a combination of both arrangements, (i.e., several crystals arranged as in Fig. 7a) set up at different angles surrounding the sample will allow optimum exploitation of the available intensity. In this case the angular separation between the individual arms will be of the order of some 15° or more and the possibility of rotating the entire detector arrangement through this angular range should be provided in order to be able to cover intermediate scattering angles if desired. The area of the

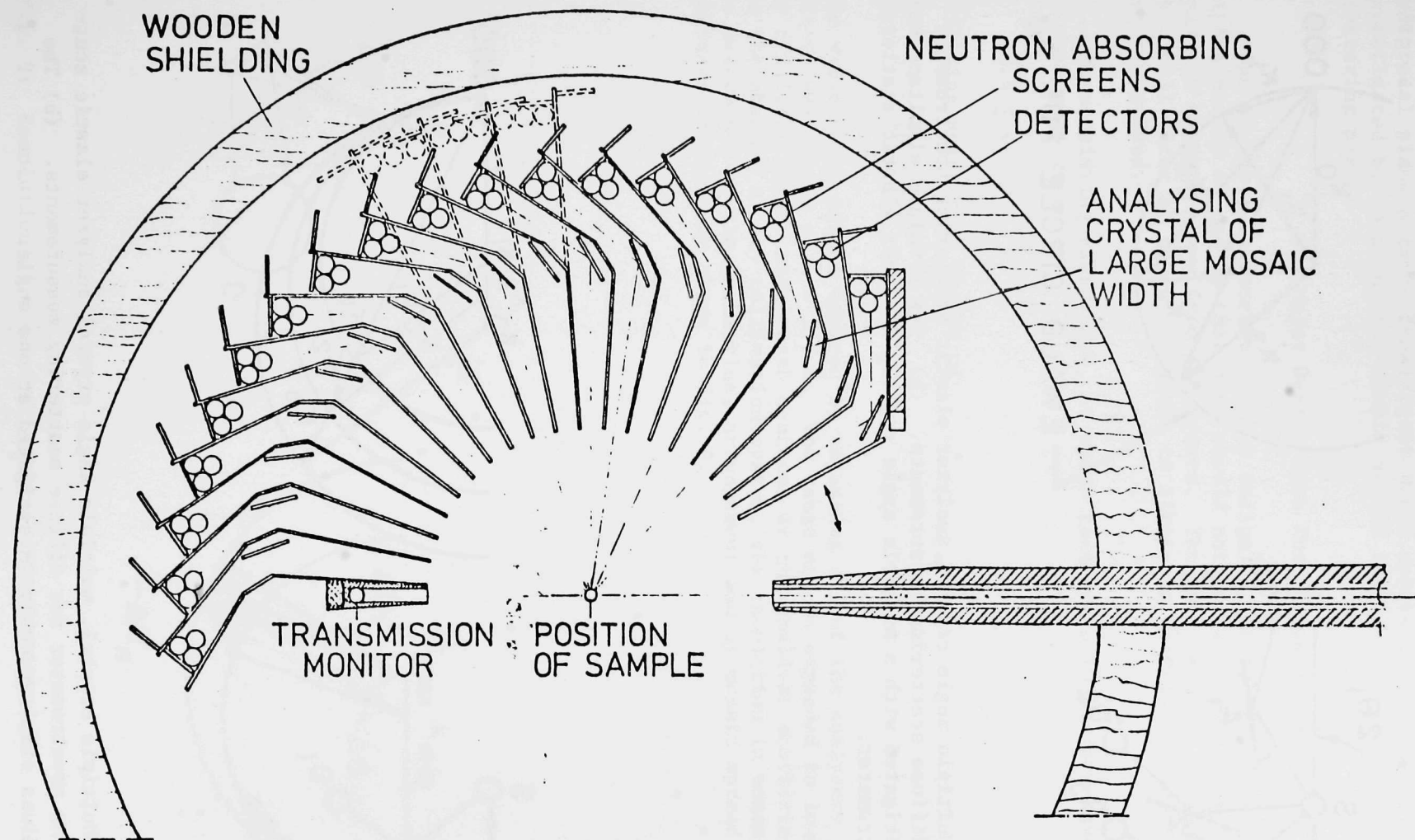


Fig. 6: Arrangement of many analyzer-detector systems around sample for simultaneous investigation of diffraction κ -values. (The array shown here corresponds to a transmitted wavelength of 3.3 \AA and is taken from Ref. 11.) The layout shows some features typical for elastic diffuse scattering such as the heavy shielding and space for a large vacuum container.

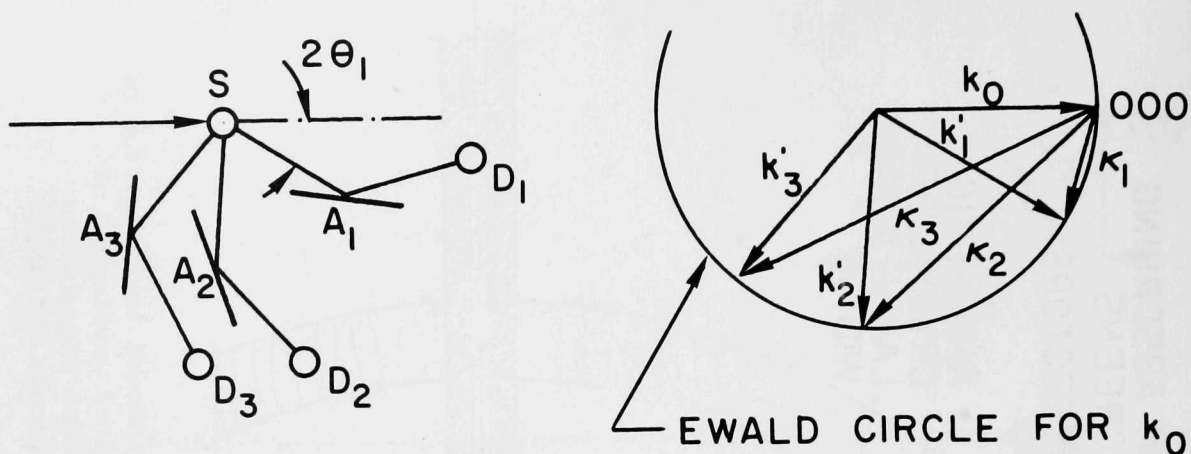


Fig. 7: (a) Multiple angle crystal analyzer elastic scattering spectrometer for diffuse scattering measurements. (b) The k -values simultaneously investigated with a multiple angle crystal analyzer elastic scattering spectrometer.

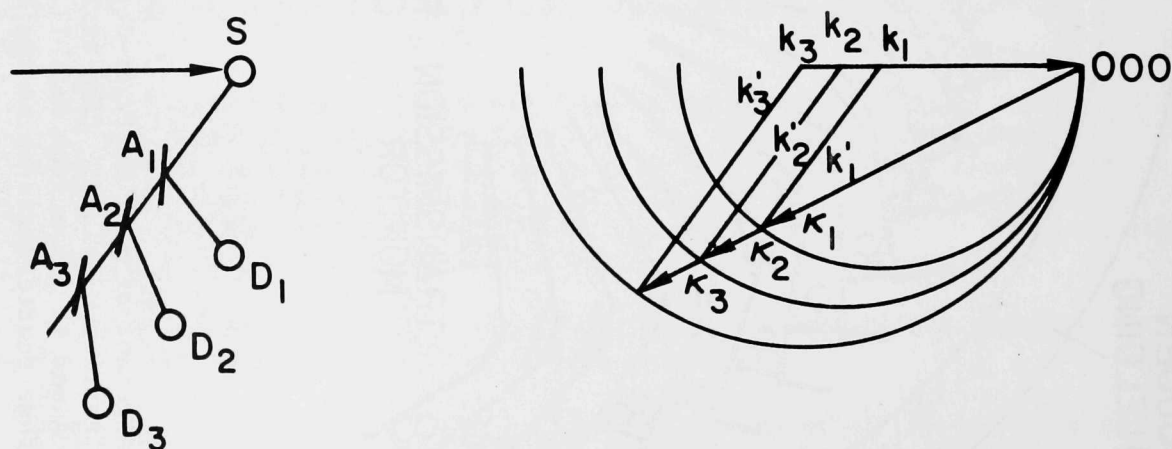


Fig. 8: (a) Multiple crystal, multiple angle crystal analyzer elastic scattering spectrometer for diffuse scattering measurements. (b) The k -values simultaneously investigated at one angle.

reciprocal plane outside the largest Ewald sphere of Fig. 8b has to be investigated by rotating the sample around its axis perpendicular to the scattering plane.

d. Major Design Data for the Instrument

One of the main concerns in the design of the spectrometers to be used with a pulsed source is that data should not be recorded during the time, a pulse is being released from the source. The intense burst of fast neutrons that will occur at these instances is likely to contribute significantly to the background.

From existing experience, it seems reasonable to perform investigations in a κ range given by

$$0.4 \leq \kappa \leq 8 \text{ \AA}^{-1}$$

utilizing scattering angles of

$$5^\circ \leq 2\theta \leq 170^\circ .$$

The whole range can be covered by adjusting all of the analyzers to a transmitted wavelength of $\lambda \approx 1.5 \text{ \AA}$. The range may be expanded to lower κ values by utilizing longer wavelength neutrons at the smallest scattering angles. In the absence of any collimation system, the resolution in momentum space is determined by the geometrical arrangement and by mosaic spread of the analyzer crystals as shown in Fig. 9.

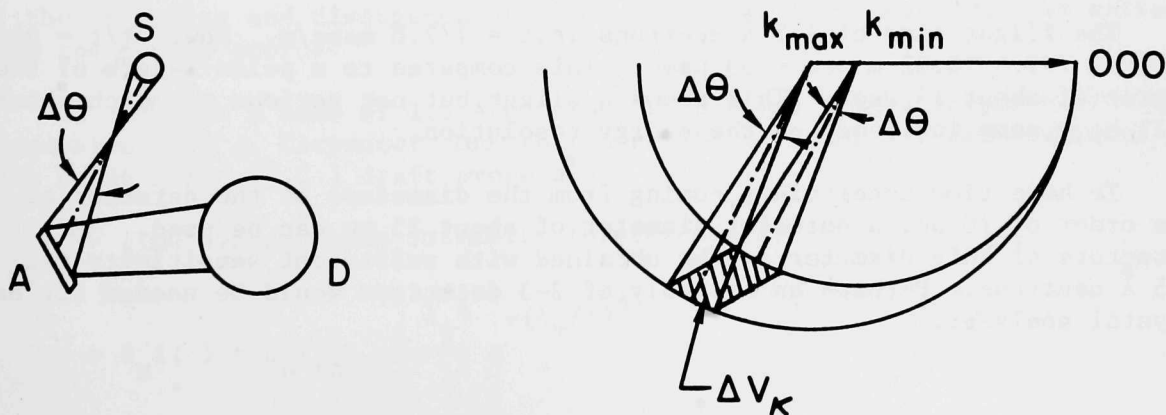


Fig. 9: Resolution of a crystal analyzer spectrometer.

$\Delta\theta$ corresponds to the effective angle under which the analyzer is seen from the sample and $\Delta\kappa$ may be estimated from the differentiated Bragg equation

$$\left| \frac{\Delta\kappa}{\kappa} \right| = \left| \frac{\Delta\lambda}{\lambda} \right| = |\operatorname{ctg} \theta \Delta\theta| + \left| \frac{\Delta\tau}{\tau} \right|. \quad (3)$$

Neglecting to a first approximation, the radial mosaic spread $\Delta\tau/\tau$ we have

$$\left| \frac{\Delta\kappa}{\kappa} \right| = \operatorname{ctg} \theta \Delta\theta. \quad (4)$$

For the case of pyrolytic graphite analyzers, we have $2d = 6.7 \text{ \AA}$, and, for $\lambda = 1.5 \text{ \AA}$, $\theta \approx 13^\circ$. A 50 mm wide crystal slab at a distance of 50 cm from the sample will, therefore, subtend an angle of $\Delta\theta_A = (50 \cdot \sin 13^\circ / 500) \approx 1.3^\circ$. Assuming a mosaic width $\Delta\theta_m = 0.5^\circ$ we have

$$\Delta\theta = \sqrt{\Delta\theta_A^2 + \Delta\theta_m^2} \approx 1.4^\circ.$$

This leads to about 5% resolution in κ , whence some coarse collimation in front of the analyzer crystal may become necessary.

To match the divergence of the beam incident on the sample roughly to that of the analyzer θ_m we obtain for a sample width of 2 cm and a source width of 10 cm a distance of

$$L = \frac{(10 + 2) \cdot 10^2}{1.3 \cdot \pi / 180} = 5.3 \text{ m}.$$

The biological shielding must be at least 5 meters thick; therefore, a distance of source to detector of 7 m seems adequate for all purposes, including the shielding of the spectrometer.

The time-of-flight resolution of the facility will be optimized if the pulse width of the source and the time-spread of the wavelength interval $\Delta\lambda$ selected are about the same magnitude. With $\Delta\lambda/\lambda \approx 0.02$, $\lambda = 1.5 \text{ \AA}$ we have $\Delta\lambda = 0.03$ at $\lambda = 1.5 \text{ \AA}$.

The flight time of 1.5 \AA neutrons is $t = 1/2.6 \text{ msec/m}$. Now, $\Delta t/t = \Delta\lambda/\lambda$ or $\Delta t = 7/2.6 \cdot 0.02 \text{ m sec} = 53 \text{ \mu sec}$. This compares to a pulse length of the source of about 14 \mu sec . This shows a slight but not serious mismatch which will have some influence on the energy resolution.

To have time uncertainty coming from the diameters of the detector of the order of 10 \mu s , a detector diameter of about 25 mm can be used. ^3He detectors of this diameter can be obtained with sufficient sensitivity for 1.5 \AA neutrons. Perhaps an assembly of 2-3 detectors would be needed for each crystal analyzer.

e. Analyzer Crystals

The performance of graphite crystals as analyzers may be estimated from the data given by Riste and Otnes.¹² One gets for the integrated reflectivity R^θ of a crystal of thickness t_0 and mosaic width β :

$$R^\theta = 0.61 (\beta \cdot Q \cdot t_0 / \sin \theta)^{1/2} \quad (5a)$$

where Q is given by

$$Q = (\lambda^3 / \sin 2\theta) \cdot 5.65 \times 10^{-3} \text{ cm}^{-1} \quad (\lambda \text{ in } \text{\AA}) \quad (5b)$$

The wavelength interval transmitted by the analyzer is

$$R^\lambda = R^\theta \cdot \lambda \cdot \text{ctg } \theta \quad (6)$$

and the maximum reflectivity is

$$R^{\max} = \frac{A}{1 + A}, \quad A = 0.94 (Q \cdot t_0 / \beta \cdot \sin \theta) \quad (7)$$

Using $\lambda = 1.5 \text{ \AA}$, $\Delta\lambda/\lambda = 0.02$, $\beta = 0.5^\circ$, $\theta = 12.94^\circ$ we obtain from Eq. (5b) a value of $Q = 0.0437 \text{ cm}^{-1}$. From $R^\lambda = 0.03$ we obtain from Eq. (6) $R^\theta = 4.6 \times 10^{-3}$ and from Eq. (5a) we calculate a minimum thickness of the crystal $t_0^{\min} = 0.034 \text{ cm}$. Crystals with $t_0 = 1.5 \text{ mm}$ are available commercially. Using this value we obtain

$$R^{\max} = 0.76$$

for the maximum reflectivity of the crystal. With thickness increased above t_0^{\min} we may use this value for the whole interval $\Delta\lambda$.

f. Intensity Estimate

To have some basis for comparing the proposed instrument at IPNS and existing steady-state neutron facilities, we have tried to obtain an estimate of the intensity and divergence of a monochromatic beam that could be extracted from the pulsed source.

We consider a beam of 1.5 \AA neutrons with a wavelength spread $\Delta\lambda/\lambda = 0.02$. Discussion with J. Carpenter led to the following numbers using moderator C (see p. 96 Table IV C.1 draft proposal).

ϕ = time average beam intensity (n/ster-sec- \AA)

$$\begin{aligned} &= S_n I(\lambda) = S_n I_{\text{ther}} \frac{1}{2} \frac{\lambda_T^4}{\lambda^5} e^{-(\lambda_T/\lambda)^2} \\ &= \frac{S_n I_{\text{ther}}}{EI(E)_{\text{1eV}}} \frac{1}{2} \frac{\lambda_T^4}{\lambda^5} e^{-(\lambda_T/\lambda)^2} EI(E)_{\text{1eV}} \end{aligned} \quad (8)$$

where S_n = source neutrons/unit time, $EI(E)_{1\text{eV}}$ (n/ster- S_n) is the value of $EI(E)$ at 1 eV, $I_{\text{the}}/EI(E)_{1\text{eV}}$ relates to the Maxwellian portion of the spectrum. With $S_n = 9 \times 10^{16}$ n/sec, $\lambda = 1.5 \text{ \AA}$, $\lambda_T = 1.57 \text{ \AA}$ (0.033 eV); $I_{\text{ther}}/EI(E)_{1\text{eV}} = 3.5$ (Table IV C.2, p. 104) and $EI(E)_{1\text{eV}} = 4.4 \times 10^{-4}$ (Table IV C.1, p. 96) we have $\phi = 1.8 \times 10^{13}$ n/ster $\text{\AA} \cdot \text{sec}$. The moderator is seen from a point distance L from the source under a solid angle

$$\Omega = \frac{10^2}{L^2 [\text{cm}^2]},$$

since the source is 10 cm on an edge.

Considering a 1 cm^2 beam at distance L from the moderator, with a $\Delta\lambda = 0.03 \text{ \AA}$ the beam intensity becomes

$$I = 1.8 \times 10^{13} \frac{10^2}{L^2} \times 0.03 = \frac{5.4 \times 10^{13}}{L^2} \quad (9)$$

Using an L of 700 cm we have then

$$\text{Time averaged intensity} \approx 1.1 \times 10^8 \frac{\text{n}}{\text{cm}^2 \text{ sec}}$$

in our wavelength interval of 0.03 \AA at $\lambda = 1.5 \text{ \AA}$.

Similar intensities may be expected for the other wavelengths close to 1.5 \AA selected if a design according to Fig. 8 is chosen. The incident flux of monochromatic neutrons on the chopper of the D7 spectrometer which is located at a cold source at ILL (Grenoble) is 2×10^7 n/cm²sec at $\lambda = 5.1 \text{ \AA}$ with $\Delta\lambda = 0.1 \text{ \AA}$ (data from Ref. 15). The intensity actually used is reduced by the duty cycle of the chopper by a factor of 1/10 to 1/20. In order to make a comparison (which will always suffer from the difference in wavelengths used) we still have to take into account the losses from the analyzer crystals in the present instrument and the number of angular positions for which data are taken simultaneously (32 at D7). The number of analyzer crystals used here is mainly a question of cost. Therefore, we make the comparison per detector used. Taking into account the reflectivity of the analyzer crystal ($R^{\text{max}} = 0.76$) and the solid angle seen by each detector ($5 \times 15/(97)^2$ at D7 and $1.2 \times 8/60^2$ here) we arrive at the following relative intensity figures

$$\text{D7: } I_{5.1 \text{ \AA}}^{\text{rel}} = 2 \times 10^7 \frac{\text{n}}{\text{cm}^2 \text{ sec}} \times 8 \times 10^{-3} \frac{\text{sterad}}{\text{det}} \times \frac{1}{20} = \underline{8 \times 10^3}$$

$$\text{Present: } I_{1.5 \text{ \AA}}^{\text{rel}} = 1.1 \times 10^8 \frac{\text{n}}{\text{cm}^2 \text{ sec}} \times 2.5 \times 10^{-3} \frac{\text{ster.}}{\text{det.}} \times 0.76 = \underline{2 \times 10^5}$$

This comparison, based on the very rough and approximate design data given here shows convincingly that investigations of the diffuse elastic scattering which take of the order of a month at the most powerful instrument now available (D7 at ILL) could be done in a matter of about a day with the facility proposed here of similar resolution. Needless to say, that with the extended κ -range and the higher intensity, problems could be attached which are impractical at the present time.

g. Summary of Special Advantages of the Pulsed Source Crystal Analyzer Spectrometer

As compared to a conventional time-of-flight spectrometer for elastic diffuse scattering there are some advantages of the instrument proposed here which deserve special mention:

i. There are several incident energies that can be used; this constitutes an immediate gain equal to the number of different energies.

ii. The resolution in κ space depends on the mosaic width of the analyzer crystal employed rather than on the $\Delta\lambda/\lambda$ of the incident beam. Therefore, this can be varied for different κ values by using different grades of graphite crystals. This may become of particular importance at κ values close to Bragg peaks.

iii. The peak flux at the pulsed source can be utilized fully.

iv. There is no rotating or fast moving parts in the instrument, which are difficult to build as well as to maintain.

v. The space requirements of the analyzer parts are small as compared to conventional TOF analyzers.

vi. Recording energy transfer spectra rather than the elastic scattering alone allows one to integrate over any quasielastic scattering properly and to judge to what degree multiphonon scattering is influencing the intensity under the elastic peak.

vii. Using relatively short wavelength neutrons reduces absorption corrections.

viii. The instrument is excellently suited also for low resolution inelastic studies to get a quick overall idea of $S(\kappa, \omega)$. Inelastic events can be recorded routinely with the elastic scattering and any new unexpected phenomena would be immediately revealed.

2. Small Angle Scattering at a Pulsed Source

Neutron small-angle scattering machines are basically very long instruments, mainly due to the large sample sizes required as a consequence of the low luminosity of the neutron sources.

With a length of the order of 40 to 80 m as in Julich and Grenoble¹³ the frame overlap of the long wavelength neutrons from different pulses

would become so serious that only quasicontinuous operations would be possible. In other words, an instrument would have to be set up, making use of a conventional low-resolution monochromator and a position sensitive detector. In this case, no specific use would be made of the pulsed nature of the source and only the time average of the flux at the wavelength chosen (typically of the order of 4 - 10 Å) would have to be considered in comparing the facility with existing ones. Design principles of such machines are well established^{13,14} and will not be considered here. However, it should be emphasized that considerable improvement in the performance of position sensitive detectors may be anticipated and, therefore, the construction of a conventional SANS machine even at time average fluxes lower than at the present high flux steady state sources may be a worthwhile effort.

In the following paragraphs we would like to discuss an alternative design which might allow one to take advantage of the pulsed source by using time-resolution and a broad wavelength spectrum to cover a wide range in κ .

Disregarding for the present the technical difficulties (which may be considerable but could probably be solved) in building the collimators necessary we provide basic design data for an instrument with the following specifications:

- 1) $5 \times 10^{-3} \text{ Å}^{-1} < \kappa < 0.5 \text{ Å}^{-1}$
- 2) $\Delta\kappa/\kappa = 0.1$
- 3) Total length $< 10 \text{ m}$ to avoid frame overlap for $\lambda < 10 \text{ Å}$ at 30 cycles/sec.

The basic design is outlined in Fig. 10. Two dimensional Soller collimators are used before and after the sample. To optimize the instrument in the sense of Ref. 13 (Eq. (10)) we have

$$\langle \delta\kappa^2 \rangle = \frac{k^2}{12} \left[\left(\frac{d_D}{L} \right)^2 + \left(\frac{d_E}{\ell} \right)^2 + d_S^2 \left(\frac{1}{L} + \frac{1}{\ell} \right)^2 + \left(\frac{\Delta\lambda}{\lambda} \right)^2 \theta^2 \right]. \quad (10)$$

In our case $L = \ell = 5 \text{ m}$

$$d_D = d_E = d_S = y \quad \text{and we take} \quad \frac{\Delta\lambda}{\lambda} \times \theta \approx \frac{y}{L},$$

so

$$\langle \delta\kappa^2 \rangle^{1/2} = k \times \sqrt{5/12} \times \frac{y}{L} = 5 \times 10^{-4} \text{ Å}^{-1} \quad (11)$$

where, for $\kappa_{\min} = \frac{2\pi}{10} \text{ Å}^{-1}$, we have

$$y = 5000 \text{ mm} \times \frac{5 \times 10^{-4}}{2\pi} \times 10 \times \sqrt{12/5} = \underline{6.16 \text{ mm}}$$

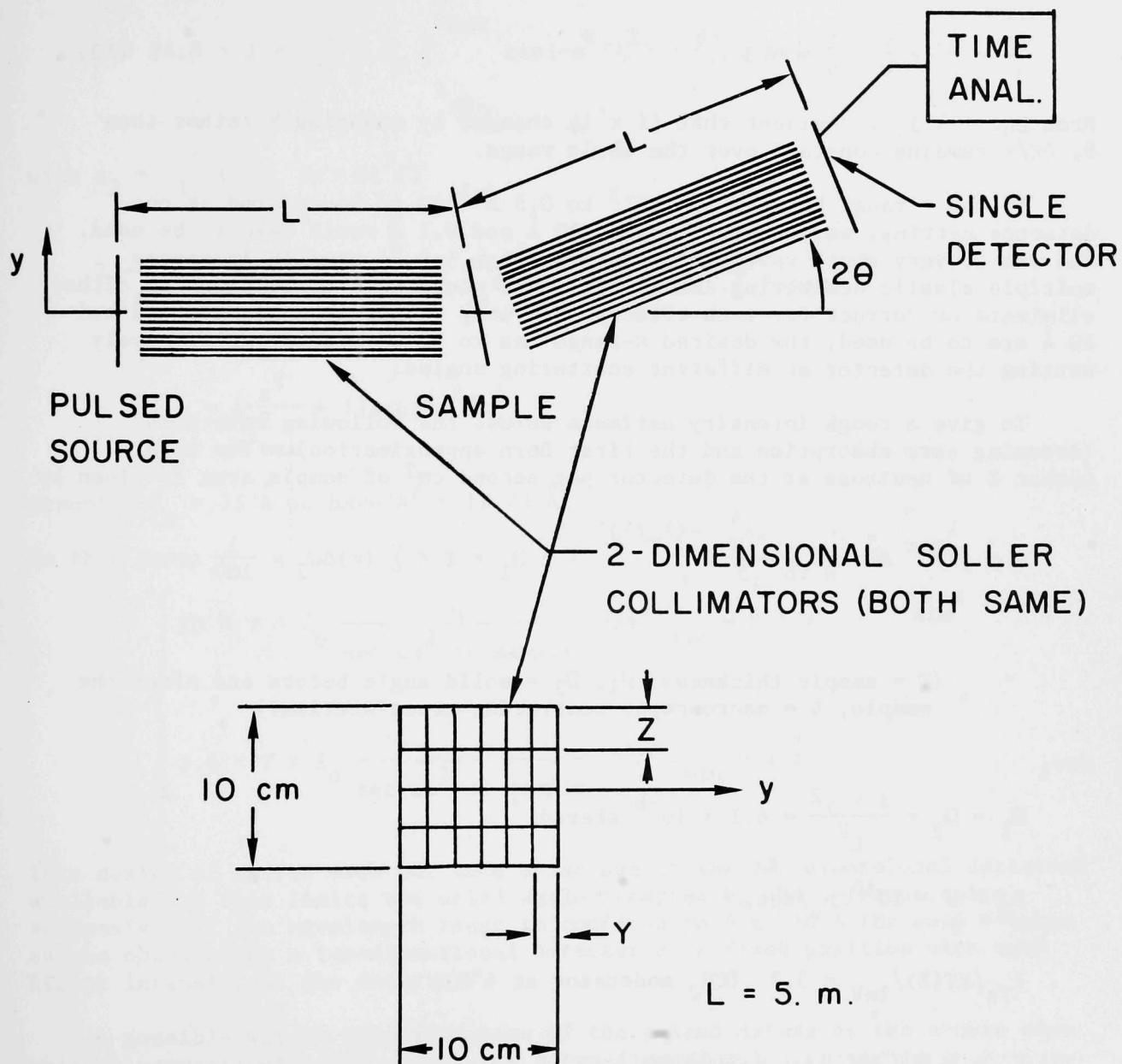


Fig. 10. Concept of a double Soller collimator SANS instrument with TOF analysis.

It seems reasonable to make $z \approx 4y \approx 25$ mm. The scattering angle for $\kappa_{\min} = 5 \times 10^{-3} \text{ \AA}$ is

$$\theta = \frac{\lambda_{\max} \times \kappa_{\min}}{4\pi} = 4 \times 10^{-3} \text{ rad.} = 0.23^\circ. \quad (12)$$

[This means that the primary beam has to be stopped by the collimator walls(!).] The time resolution needed for the detector is obtained from

$$\frac{\Delta t}{t} = \frac{\Delta \lambda}{\lambda} = \frac{y}{L} \times \frac{1}{\theta} = 0.3. \quad (13)$$

From Eq. (1) it is evident that if κ is changed by changing k rather than θ , $\Delta\kappa/\kappa$ remains constant over the whole range.

If the κ range between 5×10^{-3} to 0.5 \AA^{-1} is to be covered at one detector setting, wavelengths between 10 \AA and 0.1 \AA would have to be used. The use of very short wavelengths (λ less than $\sim 4 \text{ \AA}$) usually increases multiple elastic scattering and special care would have to be taken to either eliminate or correct for such effects. If only wavelengths between 4 \AA and 10 \AA are to be used, the desired κ -range has to be covered by successively setting the detector at different scattering angles.

To give a rough intensity estimate we use the following relations (assuming zero absorption and the first Born approximation). The total number Z of neutrons at the detector per second cm^2 of sample area is given by

$$Z = \int_{\lambda_{\min}}^{\lambda_{\max}} d\lambda S_N I_{\text{Th}} \frac{2\lambda_T^4}{\lambda^5} e^{-(\lambda_T/\lambda)^2} \times \Delta \Omega_1 \times T \times \sum (\kappa) \Delta \Omega_2 \times \frac{1}{100}.$$

(T = sample thickness, Ω_1, Ω_2 = solid angle before and after the sample, Σ = macroscopic scattering cross section.)

Now,

$$\Omega_1 = \Omega_2 = \frac{4 \times y^2}{L^2} = 6.1 \times 10^{-6} \text{ sterad},$$

$$S_N = 9 \times 10^{16} \text{ n}_s/\text{sec},$$

$$I_{\text{Th}}/EI(E)/_{\text{1eV}} = 3.2 \text{ (CH}_2 \text{ moderator at } 4^\circ\text{K)},$$

$$E_T = 0.007 \text{ eV}, \quad \lambda_T = 3.42 \text{ \AA},$$

$$EI(E)/_{\text{1eV}} = 2.3 \times 10^{-4} \text{ n/n}_s \text{ sterad}.$$

If

$$\int (\kappa) = \int_0 \times e^{-\frac{R_g^2 \times \kappa^2}{3}} = \int_0 e^{-\frac{R_g^2 (4\pi\theta)^2}{3\lambda^2}}$$

we have

$$Z = 24.3 \times 2 \times \lambda_T^4 T \Sigma_O \int_{\lambda_{\min}}^{\lambda_{\max}} \exp(-a^2/\lambda^2) \times \frac{d\lambda}{\lambda^5}, \text{ and}$$

$$\text{with } a_2 = \lambda_T^2 + (R_g 4\pi \times \theta)^2/3$$

$$Z = 24.3 (2\lambda_T^4) \times T \times \Sigma_O \times \frac{1}{2a^4} \left\{ \left[\frac{a^2}{\lambda_{\max}^2} + 1 \right] \exp(-a^2/\lambda_{\max}^2) - \left[\frac{a^2}{\lambda_{\min}^2} + 1 \right] \exp(-a^2/\lambda_{\min}^2) \right\}.$$

Assuming $R_g = 35 \text{ \AA}$ we have $a^2 = 12.73 \text{ \AA}^2$.

In this case:

$$Z = \begin{cases} 20 \times T \times \Sigma_O \frac{n}{\text{sec cm}^2 \text{ of sample}} & \text{for } \lambda_{\min} = 0.1 \text{ \AA} \\ 3.6 \times T \times \Sigma_O \frac{n}{\text{sec cm}^2 \text{ of sample}} & \text{for } \lambda_{\min} = 4 \text{ \AA} \end{cases}$$

This design of course does not take advantage of the two-dimensional detectors available and this limits the solid angle accepted by the detector quite seriously. If the wavelength range is confined to 4 to 10 Å the same κ -range can be obtained by a two-dimensional detector at a fixed position with much higher intensity at the detector.

A possible way to take advantage of the pulsed nature of the source when using a conventional SANS setup with a two-dimensional detector is as follows. A chopper which is synchronized with the pulsed source and which transmits neutrons at different times, i.e., with different wavelengths, say in the range between 4 Å and 10 Å in front of the sample (Fig. 11). The total length of the instrument is chosen in such a way as to allow no overlap from neutrons corresponding to the various pulses as indicated in the distance-versus-time diagram of Fig. 12.

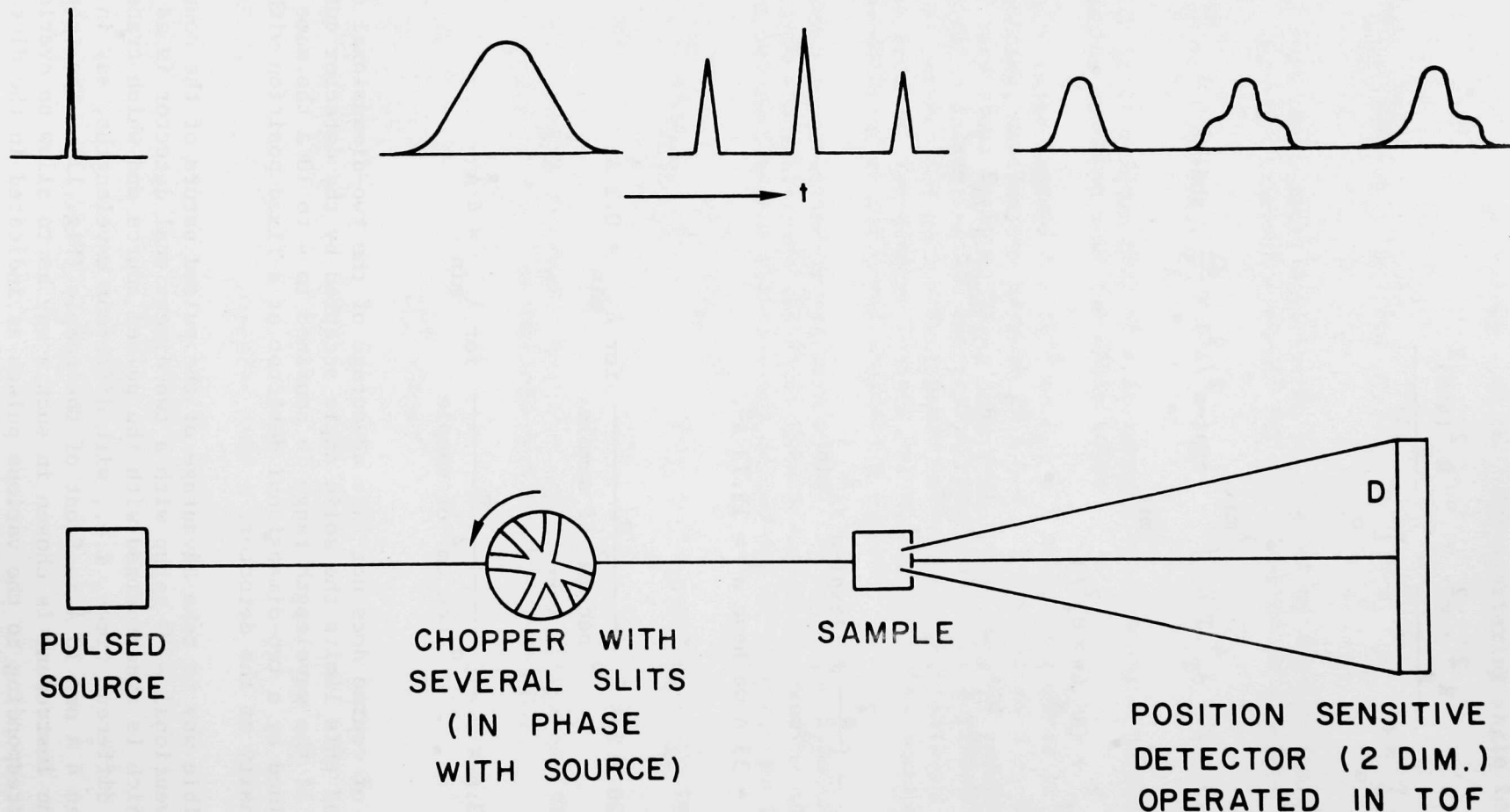


Fig. 11. SANS-instrument with phased chopper transmitting different wavelengths at different times. The spread in time (\propto wavelength) of the pulses at the various positions is indicated qualitatively.

In such an arrangement the number of wavelength transmitted per source pulse, the wavelength resolution, and the length of the instrument have to be matched to obtain optimum intensity at the detector. The obvious gain as compared to a steady state source of the same time average flux is the number of wavelengths utilized. In addition, if very long wavelength neutrons are to be employed, the number of slits in the chopper can be reduced to one, to obtain the quasicontinuously operating instrument mentioned above.



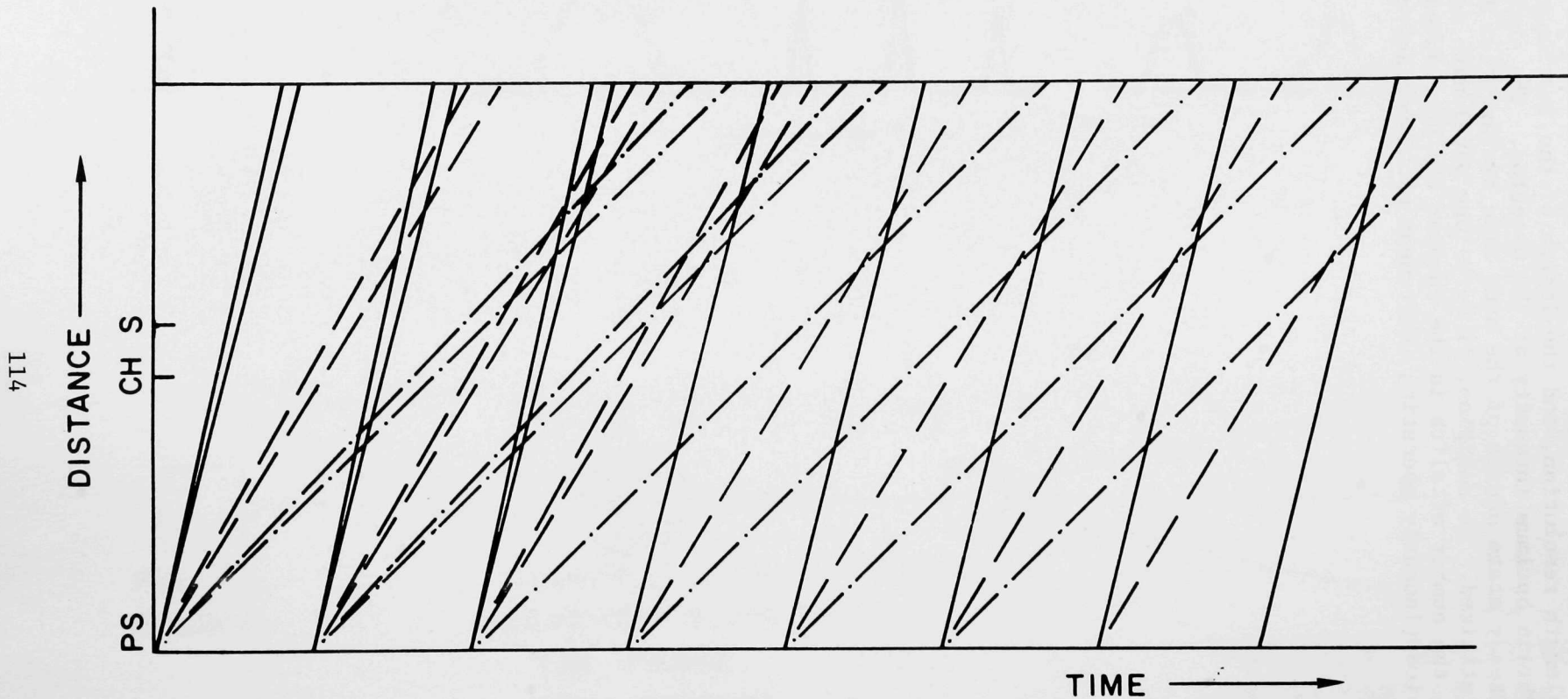


Fig. 12. Distance versus time diagram for neutrons transmitted by a 3-slit chopper. $\text{—}\text{—}\text{—}\lambda_1$, $\text{—}\text{—}\text{—}\lambda_2$, $\text{—}\cdot\text{—}\lambda_3$. The spread in wavelength is indicated for the first three source pulses. Only the trajectories of those wavelengths are shown which are transmitted by the chopper.

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V. REPORT OF THE PANEL ON DYNAMICS OF SOLIDS AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. INTRODUCTION

In an earlier survey, "Applications of a Pulsed Spallation Neutron Source" (ANL 8032), it was recognized that the most powerful applications of such a source would be the investigations for which high incident energies would be required. This panel is in complete agreement with this earlier conclusion. However, the further increase of a factor of 10 in flux from the original ZING concept to the present IPNS concept has greatly reinforced the earlier conclusions to the point where this panel finds that a general purpose, high energy-transfer, high-resolution spectrometer based on IPNS has performance characteristics, in the appropriate energy range, which are one to two orders of magnitude better than any spectrometer operating at a steady state source.

In the next section, a brief description of some of the possible IPNS spectrometers for inelastic scattering is given, together with some estimated performance characteristics. In Sec. C, we list a number of problem areas to which the IPNS can be applied and give a list of several of them for which the IPNS seems particularly suited.

B. Instruments

The basic form of most of these spectrometers is indicated schematically below.

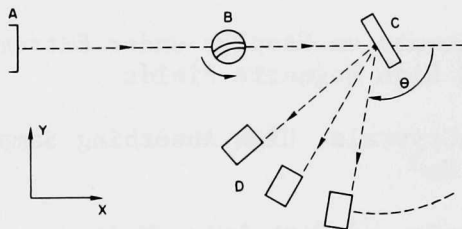


FIGURE 1
DIRECT GEOMETRY CHOPPER SPECTROMETER
AT A PULSED NEUTRON SOURCE

A represents the moderator (10 cm \times 10 cm in area) which is, for our purposes, the pulsed neutron source. B represents a multi-slit chopper, C the sample, and D one of a multi-detector array over a wide range of scattering angles (5° to 150°). If necessary, an extra chopper might be placed between A and B to sharpen the source pulse, and for some applications B itself might be replaced by a correlation chopper.

Let d_1 , d_2 , d_3 be the distances AB, BC, CD, respectively. For the general purpose time-of-flight spectrometer (energy transfers up to 150 meV), typical values of these might be $d_1 = 10$ m, $d_2 = 0.2$ m, $d_3 = 2.5$ m. We call this spectrometer I. For a high energy spectrometer (energy transfers between 150 and 1500 meV), typical values might be $d_1 = 40.0$ m, $d_2 = 0.2$ m, $d_3 = 5.0$ m, with perhaps fewer detectors, owing to the cost of the increased detector area required. We refer to this instrument as spectrometer II. A third type

of spectrometer for use with neutrons in the 3-7 eV range would dispense with the chopper and use a ^{238}U resonance analyzer. We shall refer to this instrument as spectrometer III.

(We assume here that the detector area can be "matched" to the K-resolution required by hooking several detectors together if necessary.)

The counting rate in the detector for a particular process (integrated over all energy transfers involved in that process, e.g., the integrated count-rate in a one-phonon peak) is given by

$$T = n \ 2 \ \phi_o \ e^{-E_o/E_t} (E_o/E_t)^2 \frac{1}{v_o} \Delta v_o \Delta\Omega_o \Delta\Omega_s N_s \left(\frac{d\sigma}{d\Omega}\right) \eta \quad (1)$$

where n is the number of neutron pulses/sec

$$(\text{= } 60 \text{ sec}^{-1} \text{ for IPNS})$$

$$\phi_o = 4.76 \times 10^{12} \text{ neut/steradian/eV/pulse}$$

$$E_o = \text{Incident energy (in meV)}$$

$$E_t = \text{Effective moderator temperature (in meV)} \\ (\text{= } 35 \text{ meV})$$

$$\Delta v_o = \text{Incident neutron velocity spread transmitted by chopper (cm/sec)} \\ (\text{see Eq. (2A), below})$$

$$v_o = \text{Incident neutron velocity (cm/sec)}$$

$$\Delta\Omega_o = \text{Solid angle subtended by } 1 \text{ cm}^2 \text{ of sample at source}$$

$$\Delta\Omega_s = \text{Solid angle subtended at sample by detector}$$

$$N_s = \text{Number of atoms in sample}$$

$$A_s = \text{Area of sample in beam (cm}^2\text{)}$$

$$\frac{d\sigma}{d\Omega} = \text{Differential cross-section for process (integrated over scattered energy) in cm}^2\text{/steradian}$$

$$\eta = \text{Detector efficiency} \simeq 1.$$

For our purposes, we have assumed $N_s = 10^{23}$, $A_s = 1 \text{ cm}^2$, $\left(\frac{d\sigma}{d\Omega}\right) = 1 \text{ barn/steradian}$ corresponding roughly to a 10% scatterer. For incident neutron energies greater than 150 meV, the factor

$$\phi_o e^{-E_o/E_t} \left(\frac{E_o}{E_t}\right)^2$$

should be replaced by $\phi'_o = 3 \times 10^{13} \text{ neut/steradian/eV/pulse}$ to reproduce the $(1/E)$ dependence of the source spectrum in this region.

The energy resolution is given by

$$\Delta E = m \left[\frac{1}{d_1^2} (v_o^3 + v_s^3 \frac{d_2}{d_1})^2 T_1^2 + \frac{1}{d_1^2} (v_o^3 + v_s^3 \frac{d_1 + d_2}{d_3})^2 (T_2^2 + \overline{\Delta \alpha_o^2} / \omega_c^2) + \frac{v_s^4}{d_3^2} \overline{\Delta d_3^2} \right]^{1/2} \quad (2)$$

while

$$\Delta v_o = \frac{v_o^2}{d_1} (T_1^2 + T_2^2 + \overline{\Delta \alpha_o^2} / \omega_c^2)^{1/2} \quad (2A)$$

where m is the neutron mass, v_s is the scattered neutron velocity, $\Delta \alpha_o$ is the incident-beam horizontal divergence, T_1 is the pulse-width at the source (obtained as a function of E_o from ANL 8032), T_2 the chopper on-time, and ω_c is the angular speed of the chopper. Δd_3 is the active thickness of the detector (including the effect of sample thickness), taken here as 0.6 cm.

The resolutions in K_x , K_y , K_z (normal to plane of scattering, see Fig. 1), are given by:

$$\begin{aligned} \Delta K_x = \frac{m}{h} & \left[\frac{1}{d_1^2} (v_o^2 - v_s^2 \cos \theta \frac{d_2}{d_3})^2 T_1^2 \right. \\ & + \frac{1}{d_1^2} (v_o^2 - v_s^2 \cos \theta \frac{d_1 + d_2}{d_3})^2 T_2^2 \\ & + \frac{1}{d_1^2 \omega_c^2} (v_o^2 - v_s^2 \frac{d_1 + d_2}{d_3} \cos \theta)^2 \overline{\Delta \alpha_o^2} \\ & \left. + v_s^2 \sin^2 \theta \overline{\Delta \alpha_s^2} + \frac{v_s^2}{d_3^2} \cos^2 \theta \overline{\Delta d_3^2} \right]^{1/2} \quad (3) \end{aligned}$$

$$\begin{aligned}
\Delta K_y = \frac{m}{h} & \left[\frac{1}{d_1^2} v_s^4 \frac{d_2^2}{d_3^2} \sin^2 \theta T_1^2 \right. \\
& + \frac{v_s^4}{d_3^2} \left(1 + \frac{d_2}{d_1} \right)^2 \sin^2 \theta T_2^2 \\
& + \left(v_o + \frac{v_s^2 \sin \theta}{d_3 \omega_c} \frac{d_1 + d_2}{d_1} \right)^2 \overline{\Delta \alpha_o^2} \\
& \left. + v_s^2 \cos^2 \theta \overline{\Delta \alpha_s^2} + \frac{v_s^2}{d_3^2} \sin^2 \theta \overline{\Delta d_3^2} \right]^{1/2} \quad (4)
\end{aligned}$$

$$\Delta K_z = \frac{m}{h} \left[v_o^2 \overline{\Delta \beta_o^2} + v_s^2 \overline{\Delta \beta_s^2} \right]^{1/2} \quad (5)$$

where $\Delta \alpha_s$ is the horizontal divergence of the scattered beam, $\Delta \beta_o$ the vertical divergence of the incident beam, $\Delta \beta_s$ that of the scattered beam, θ and θ_o is the angle of scattering (see Fig. 1).

We present below estimated count-rates and resolutions for spectrometers I and II for selected energy and momentum transfers, for our assumed 10% scatterer.

1. General Purpose Spectrometer

1. Spectrometer I: $d_1 = 10.0$ m, $d_2 = 0.2$ m, $d_3 = 2.5$ m
detector area 25 cm (hor) by 22.5 cm (vert)

(1) $E_o = 150$ meV, $E^1 = 50$ meV

1 μ sec chopper at 600 Hz

Resolution: $\Delta E = 2.3$ meV

$$\Delta K_{x,y} \sim 0.09 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.13 \text{ \AA}^{-1}$$

Count rate: 700/sec

K range: $3.6 \text{ \AA}^{-1} (5^\circ)$ to $13 \text{ \AA}^{-1} (150^\circ)$

(2) $E_o = 100$ meV, $E^1 = 50$ meV

1 μ sec chopper at 600 Hz

Resolution: $\Delta E = 0.80 \text{ meV}$

$$\Delta K_{x,y} \sim 0.08 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.13 \text{ \AA}^{-1}$$

Count rate: 640/sec

K range: 2.1 \AA^{-1} to 11.5 \AA^{-1}

(3) $E_0 = 20 \text{ meV}$, $E^1 = 5 \text{ meV}$

10 μsec chopper at 200 Hz

Resolution: $\Delta E = 0.3 \text{ meV}$

$$\Delta K_{x,y} = 0.04 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.04 \text{ \AA}^{-1}$$

Count rate: 470/sec

K range: 1.6 \AA^{-1} to 4.5 \AA^{-1}

Note that $\left(\frac{d\sigma}{d\Omega}\right)$ for a typical one-phonon scattering process for down-scattering is roughly given by

$$\frac{d\sigma}{d\Omega} = \left(\frac{E^1}{E_0}\right)^{1/2} \frac{\sigma_{\text{coh}}}{4\pi} \left(\frac{\hbar^2 Q^2}{2M}\right) \left(\frac{1}{\hbar\omega}\right) e^{-2W}$$

where ω is the phonon frequency and \bar{M} is an effective mass for the mode.

If we assume a mass of $\sim 60 \text{ a.m.u.}$, a K of about 10 \AA^{-1} , $\hbar\omega = 100 \text{ meV}$, $\sigma_{\text{coh}} = 5 \text{ barns}$, and $e^{-2W} \sim 1$, we obtain for $E_0 = 150 \text{ meV}$, $E^1 = 50 \text{ meV}$,

$$\frac{d\sigma}{d\Omega} \sim 8 \times 10^{-3} \text{ barns/steradian,}$$

so that the count rate in the one-phonon peak would be given by 8×10^{-3} times the count rates given above, which comes out to be of the order of several counts per second for a relatively small sample and good resolution.

2. High Energy Spectrometer

2. Spectrometer II: $d_1 = 40 \text{ m}$, $d_2 = 0.2 \text{ m}$, $d_3 = 5 \text{ m}$
detector area $12.5 \text{ cm (hor) by } 22.5 \text{ cm (vert)}$
1 μs chopper at 600 Hz

(1) $E_0 = 1500 \text{ meV}$, $E^1 = 1250 \text{ meV}$, $K = 2.6 \text{ \AA}^{-1}$ ($\theta = 2.5^\circ$)

Resolution: $\Delta E = 13 \text{ meV}$

$$\Delta K_{x,y} \sim 0.13 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.3 \text{ \AA}^{-1}$$

Count rate: 55/sec

$$(2) E_0 = 1500 \text{ meV}, E^1 = 500 \text{ meV}, K = 11.4 \text{ \AA}^{-1} (\theta = 2.5^\circ)$$

Resolution: $\Delta E = 10 \text{ meV}$

$$\Delta K_{x,y} \sim 0.09 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.20 \text{ \AA}^{-1}$$

Count rate: 55/sec

$$(3) E_0 = 500 \text{ meV}, E^1 = 300 \text{ meV}, K = 3.6 \text{ \AA}^{-1} (\theta = 2.5^\circ)$$

Resolution: $\Delta E = 2.2 \text{ meV}$

$$\Delta K_{x,y} \sim 0.05 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.16 \text{ \AA}^{-1}$$

Count rate: 32/sec

$$(4) E_0 = 2000 \text{ meV}, E^1 = 500 \text{ meV}, K = 16 \text{ \AA}^{-1}, (\theta = 2.5^\circ)$$

Resolution: $\Delta E = 15 \text{ meV}$

$$\Delta K_{x,y} \sim 0.11 \text{ \AA}^{-1}$$

$$\Delta K_z = 0.20 \text{ \AA}^{-1}$$

Count rate: 64/sec

C. NEW RESEARCH AREAS

1. Experiments involving Energies up to 150 meV

a. Measurements in Disordered and Polycrystalline Materials

As is evident from the figures given in Sec. II, the high intensities and good resolutions available with spectrometer I in the 50-150 meV energy transfer region are far superior to those obtainable with present steady-state sources. Unfortunately, the scattering cannot be conveniently done for selected momentum transfers. However, for experiments where preferred wave-vector selection is not essential, and where the multidetector arrangement may be usefully exploited, the IPNS spectrometer would be unsurpassed by any other. Examples of such experiments are:

i. Measurements of $S(K, \omega)$ for Amorphous Materials

Such experiments are being attempted nowadays but are difficult to do even on the present high-flux reactors. Spin-wave excitations in amorphous ferromagnets have recently been found to exhibit interesting roton-like

behavior.¹ Such measurements can be performed on spectrometer I for energy transfers up to 100 meV, and for K values $< 3 \text{ \AA}^{-1}$ (corresponding roughly to the "first zone") with excellent resolution and counting rates. If necessary, even at these small values of K, phonons may be discriminated against by the use of polarized neutrons (using a pulsed polarized proton filter in the incident beam).

ii. Measurements of $S(K, \omega)$ from Hydrogen in Metallic Hydrides
with Large Concentrations of Hydrogen Vacancies

Existing experiments² have already shown that in such systems, the one-phonon density of states for the optic modes calculated via dispersion curves obtained from the centroids of the broad "one-phonon" peaks in coherent scattering measurements³ are not sufficient for a complete picture of the dynamics of these disordered systems. Rather, a complete knowledge of $S(K, \omega)$ is required. Existing experience² with the ZING-P runs indicates that such information may be obtained with spectrometer I on IPNS with excellent ($< 1 \text{ meV}$) resolution in a matter of a few minutes in some cases, to a few hours in others.

iii. Measurements of the Phonon Spectra of Superconducting Materials

By working on cooled samples (to minimize multi-phonon scattering) in K ranges from $9\text{--}11 \text{ \AA}^{-1}$ and using averaging over several detectors, the "incoherent approximation" for powder samples could be made reasonably valid. Of course, for non-Bravais lattices, one would observe not the true frequency spectrum, but rather an eigenvector-weighted frequency spectrum if the atoms had different σ/M ratios. A rough idea of the count rates may be obtained for a measurement of the phonon spectrum of V_3Si (cut-off frequency $\sim 50 \text{ meV}$) with an incident energy of 100 meV, an average energy resolution of 1 meV, and wavevector resolution of 0.2 \AA^{-1} per detector, for a 1 cc sample. This comes out to be at an average value of about 2 cts/sec per meV energy interval per detector over the whole system, or 1% accuracy in about 90 minutes per detector, using spectrometer I.

With the excellent resolution available, it should be possible to measure fairly rapidly the characteristic features of the spectrum as well as temperature or pressure shifts of the spectrum. The temperature shifts of the phonon spectra of strong-coupled superconductors such as the β -tungsten materials, for instance, are believed to be related to the change in the electron screening of the electron-phonon interaction.⁴ The pressure effects might be interesting to observe in conjunction with very high pressure pulses applied in phase with the neutron bursts, as discussed below.

b. Experiments on Samples under Extreme Conditions-Pulsed
High Magnetic Fields

The pulsed nature of the source may be effectively exploited by pulsing a sample to extreme conditions (e.g., high magnetic fields) at the frequency (or submultiple thereof) of the source. A measurement of the scattering function $S(K, \omega)$ of the sample under the applied field can then be measured provided the applied conditions persist over at least 20 microseconds or so. For example, it should be possible to pulse a 250 kOe magnetic field on

the sample 60 times a second, or to pump electron or phonon levels in solids using infra-red laser pulses and perform excited state spectroscopy.

Another possibility is to observe transient phenomena by phasing the pulsed sample condition relative to the arrival of the neutron pulse at the sample. Examples of such experiments are the study of density fluctuations in the wake of a shock wave; pumping excited state levels in solids and observing the energy redistribute itself between the other levels, studies of atom migration in superionic conductors on applying large electric fields; and studies of spinodal decomposition in systems near a critical point. Domain nucleation and growth near magnetic critical points could also be studied, particularly with the availability of a small angle scattering spectrometer (not described here) on the IPNS facility.

In order to usefully observe such phenomena, however, the characteristic time constants must be between microseconds and milliseconds. It is at present not clear how many such interesting transient phenomena are suitable for study in this manner.

c. Small Crystals, Highly Absorbing Samples, e.g., He^3

There is also a large class of important experiments where the greater thermal flux of the IPNS facility would make it worthwhile to sacrifice the convenience of the "Constant Q" mode of neutron spectroscopy, such as experiments involving very small single crystals, or highly absorbing nuclei such as He^3 . For example, one may attempt to study the very weak spin scattering from dimerized molecules of organic charge transfer salts, such as excitations from the singlet to the triplet spin states which are believed to form on the molecules, or to study the coupling between the spins and the phonons or other lattice excitations.

d. Dispersion of High-Lying Modes

Finally, another promising area of application is in the area of studying the dispersion of high-lying modes associated with intramolecular vibrations in molecular crystals, where the millivolt resolution at large energy transfers of the order of 100 millivolts may be essential. A very similar problem is measurement of dispersion of the optical modes associated with hydrogen in a metal hydride. This may be studied effectively from deuterated single crystal samples with spectrometer II, thus yielding important information regarding effective hydrogen-hydrogen interactions in these materials. Such measurements have recently been successfully made at a high flux reactor on $\text{PdD}_{0.63}$ but we may envisage more difficult problems of the same type being attempted on the IPNS facility.

2. Experiments involving Energy Transfers from 0.15 eV to 5 eV

The prospect of the inelastic scattering experiments in the above energy range with the IPNS is one of the most exciting areas which could be opened up with such a facility. It is also a relatively unknown area in which actual experience will undoubtedly open up many new possibilities. Some possible problems considered by this panel are listed below.

a. Electronic Level Spectroscopy

Electronic level spectroscopy, where the technique would in a sense be complementary to optical spectroscopy and electron-scattering experiments. Unlike the former, it could be performed over a wide range of momentum transfers, and it would also be much freer of multiple scattering and surface effects than the latter. The matrix elements for such a scattering process are of the form

$$\langle \psi_{k\uparrow} | S_+ e^{-i\vec{q}\cdot\vec{r}} | \psi_{k+q\downarrow} \rangle \quad (\text{spin flip scattering})$$

and

$$\langle \psi_{k\uparrow} | \left\{ \frac{P}{2m} \vec{x} e^{-i\vec{q}\cdot\vec{r}} + e^{-i\vec{q}\cdot\vec{r}} \frac{P}{2m} \vec{x} \right\} | \psi_{k+q\uparrow} \rangle \quad (\text{orbital scattering})$$

in the one electron Bloch picture. In the tight-binding approximation, for small \vec{q} , the former gives electric-dipole allowed transitions while the latter gives magnetic dipole allowed transitions. It should be noted that the matrix element for spin flip scattering is, for paramagnetic materials (in the absence of spin-orbit coupling) identical to that for electron scattering. However, the absence of the $1/q^4$ term in the cross section enables one to look at regions of much larger momentum transfer (but smaller energy transfer) than electron scattering. It should be noted that apart from the above matrix elements, which essentially behave as form factors, the cross sections for such processes can be quite large, i.e., of the order of millibarns, and comparable to those for one-phonon scattering. The kinematics of the neutron scattering process make it difficult to obtain small momentum transfers for such energy transfers in this range, so there is some question as to whether the form factors make the intensity for such processes inadmissibly small for momentum transfers obtainable. For 3d metals, the spin form factor is negligible by $Q \sim 8 \text{ \AA}^{-1}$, but the orbital form factor is only down to 0.1. For many rare-earth metals and actinides, it is down to this value at K out to $10\text{--}12 \text{ \AA}^{-1}$. For scattering between bonding and anti-bonding states across the gap of a semiconductor, the orbital form factor is not negligibly small in such K ranges. We thus conclude that such experiments with energy transfers up to 1 eV are in fact quite feasible using spectrometer II with estimated count rates of ~ 0.1 ct/sec per resolution element which appear to be quite impressive. The resolution obtainable would appear to be < 30 meV which is to be compared with resolution of ~ 100 meV currently obtainable in electron scattering experiments. For higher energy transfers, spectrometer III could be used. This technique appears to be capable of being used for a vast range of problems. One interesting application would be to magnetic insulators, and another to studies of excitations between multiplet levels in metallic rare-earth systems, particularly if one of these levels lay close to or hybridized with bands of the Fermi level.

b. Neutron Scattering Measurements of the Ground State Momentum Distributions in Materials

When neutrons of sufficiently high momentum transfer Q are scattered from materials the scattering may be thought of as taking place from the individual atoms and collective effects are not important in the scattering process. For most materials K must be very large to approximate this condition and only in liquid ^4He is the K value needed sufficiently low that measurements can be made with existing facilities.⁵⁻⁷ Fortunately ^4He is an extremely interesting system to study since it is expected that as the temperature is lowered through the λ point of the ^4He atoms will condense into a zero momentum in which K was about 15 \AA^{-1} ,⁵⁻⁷ but it would be very desirable to do the experiment at a higher K momentum transfer in case that 15 \AA^{-1} is not a high enough momentum transfer to be in the single particle regime. Other materials could be examined also at still higher momentum transfers.

The scattering law $S(K, \omega)$ for a material can be written directly in terms of the cross section per unit energy per unit solid angle in the following manner

$$\frac{d^2\sigma}{d\Omega d\omega} = C \frac{k_f}{k_i} S(K, \omega) \quad (1)$$

where k_f and k_i are the final and incident wavevectors, respectively, and C is a constant. When the momentum transfer is high enough that the neutron only scatters from individual atoms the impulse approximation is valid and $S(K, \omega)$ is given by

$$S(K, \omega) = \sum_p n_p \delta\left(\omega - \frac{\hbar K^2}{2M} - \frac{K \cdot p}{M}\right) \quad (2)$$

where p is the momentum of the atom before the scattering, and M is its mass. If we differentiate Eq. (2) we obtain the relation

$$pn_p \Big|_{p_{\min}} = \frac{4\pi K^2 \rho}{M^2} \frac{\partial S(K, \omega)}{\partial \omega} \quad (3)$$

where $p_{\min} = [\omega - \hbar K^2/2M](M/K)$ and ρ is the density of the material. The momentum distribution can thus be obtained directly from Eq. (3) if $S(K, \omega)$ has been determined. Usually in a time-of-flight experiment the measurements of $S(K, \omega)$ are made at constant angle rather than constant Q and relation 3 must be modified some what. The modifications necessary are discussed in Ref. 7.

If the IPNS neutron source were available, a much improved experiment on ^4He could be done in a very straightforward manner. A suggested incoming neutron energy is about 1.5 eV, which would give a momentum transfer of about 40 \AA^{-1} if a scattering angle of about 160° were used. The suggested experi-

mental resolution is about 10 millivolts which could give about 6 resolution elements across the halfwidth of $S(K, \omega)$. The zero momentum condensate should be readily observable with this resolution. The counting rate in the experiment would be about 10 cts/min per time-of-flight channel if about 50 channels were spread over the peak in $S(K, \omega)$. This is obviously a rather easy experiment since such a high source flux is available and measurements of the momentum distribution could be made against variables such as pressure or temperature. It might be desirable to perform a still higher momentum transfer experiment. This could obviously be accomplished since the 40 \AA^{-1} experiment is rather easy. At higher momentum transfers it may be difficult to make choppers with sufficiently high energy resolution and use may be made of resonant absorbers like ^{238}U to determining the scattered neutron energy.

Measurements of n_p would also be of great interest in other materials although of course no condensate would exist. Suggested candidates might be lithium or neon. Momentum transfers in the range of 50 \AA^{-1} to 200 \AA^{-1} might be needed and Gibbs and Harling⁸ have indicated how low K experiments of this type can be used to provide useful information about n_p and the interatomic potential of condensed systems in general. From n_p other quantities of interest such as the single particle density matrix are easily obtainable. The high K experiments are certainly not possible at existing steady state sources and a high flux pulsed source appears to be ideal.

c. High Energy Optic Modes

Instrument II could also be used to study high energy optic modes (and higher harmonics of these modes in ionic metal hydrides such as TiH_2 , ZrH_2 , CeH_{2+x} , etc.). The information obtained from such measurements could then be used to establish the potential in these systems for which single crystals will probably never be available. The extremely good energy resolution and small momentum transfers associated with this machine are required to make the analysis of the results meaningful. Another possible class of hydride experiments would be very dilute systems with H/metal ratios less than 10^{-3} . Here one can study both the parameters of the potential well for an isolated hydrogen atom in a metal by studying the frequencies, lineshapes and intensities of the localized mode scattering and its higher harmonics, and also the tendency of the hydrogens to form clusters as the concentration increases with consequent broadening of the local mode peak. If it is possible to relax some of the resolution requirements to say 9 meV for some measurements, then one could gain perhaps factors of 100 in intensity. This would then make the counting times of the order of minutes rather than days and would allow rapid accumulation of data as a function of some variable, e.g., the local mode frequency of low concentration hydrides as a function of temperature.

D. CONCLUSIONS

The IPNS facility in conjunction with the inelastic scattering spectrometers discussed here, we find to be in many ways a unique and impressive potential tool for solid state research in several areas. We have attempted to discuss some of these. In other areas, such as quasielastic scattering studies of diffusion in solids, the facility does not have a clear competitive advantage over the potential of existing high-flux reactors, but is probably still capable of producing good results. Another potential we have

not explored in detail in this respect is the capability of gathering rapidly large amounts of data on phonon spectra of complex or low-symmetry crystals over large areas of the Brillouin zone and the consequent impact on our knowledge of atom-atom potentials in molecular crystals, for example.

Further improvement of the above spectrometers may also come with the use of a correlation-chopper to enable one to measure scattering from a wide range of incident energies simultaneously, although the application is undoubtedly complicated and as yet untested. A mechanical disk-type correlation chopper presents problems for incident energies above 200 meV, and one may have to await technical developments such as that of spin flip chopping of polarized white neutron beams with a pseudorandom sequence. It may also be possible to use a correlation chopper for simultaneous studies of transient phenomena at different times by sweeping the phases between the neutron pulse, sample and the chopper.

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VI. REPORT OF THE PANEL ON LIQUIDS, GLASSES AND GASSES AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. PRESENT STATE OF THE FIELD

It is fair to say that during the last ten years with the advent of precision scattering measurements together with molecular dynamics calculations, our understanding of the liquid state has been pushed significantly forward. But, we are still far from our goal of having a satisfactory microscopic theory of the liquid state. For this we need good measurements by a variety of techniques and extensions to the theory. In fact the considerable progress which was made in liquid state physics over the past decade was possible through the combination of various data and methods together. Further progress probably depends upon continuing this combined approach, and thus we hope that all scattering methods, PVT measurements, computer simulation and basic theory move in parallel. For this reason we give in Appendix I a summary of several other experimental techniques.

In the neutron scattering field we look forward to increasingly precise and more extensive measurements of the static and dynamic structure factors. The source improvements allow, in principle, for great improvements in the data to be made and some examples will be described below. But, this will depend upon our ability to build more stable and precise spectrometers and to construct reliable samples for extensive regions of phase space.

Our theoretical understanding of the static properties of model fluids with repulsive interactions is now fairly satisfactory. Once we add an attractive or long range part to the potential the small Q behavior of the structure factor $S(Q)$ becomes complicated. This is evident, for example, near the critical point and reflects a growth of correlations between particles in the system. We have a satisfactory understanding of the dynamical behavior for simple fluids in several regimes. For small k and ω we know the general structure of $S(Q, \omega)$ for wide ranges of Q and ω using kinetic theory. We also have simple microscopic theories for dense systems which agree with the general features of the measured $S(Q, \omega)$. Such theories take into account certain static structural correlations and the Boltzmann-like binary collision processes (which introduce one relaxation time). This naive theory cannot be quite right. There is a great deal of evidence from work in computer molecular dynamics and from theoretical work (which lead to the so-called divergence of transport coefficients), for the existence of a second relaxation time corresponding to correlated recollision processes in the fluid. This second relaxation time involves correlations persisting for times as long as 10-20 collision times. Heretofore neutron experiments have not been precise enough to extract information about this second relaxation time (i.e., deconvolute it from the effects of the first relaxation time). It would be highly desirable to be able to obtain the precision necessary to measure these effects,

Regarding our understanding of the dynamical behavior of elementary liquid metals, one would like to know the precise form of the interionic potential. The present theories are based on our knowledge of the pseudopotential, dielectric screening and the use of the second order perturbation theory--all of which are somewhat in question.

Further complication occurs in binary mixtures. Particularly interesting are the fused salts (e.g., LiF, NaCl) where the interaction is only partly understood. Here again we would like to study the partial structure factors

and the individual and cross-response functions. Understanding binary mixtures we can go to more complex liquids but very little is to be gained from the basic standpoint until we have acquired a good understanding of the simpler systems.

As regards quantum liquids, for ^3He a great deal remains to be done and for ^4He superfluid experiments in the large momentum transfer region may shed light on the question of the existence of the condensed state--a problem of fundamental interest. For liquid He I and for H_2 broader studies are needed.

In these more complex systems the interaction between experiment and computer molecular dynamics will be modified from that in simple fluids. In quantum fluids computer molecular dynamics will not be directly applicable initially because of the serious complications of quantum statistics. In molecular systems the uncertainties in the interaction potential severely limit the degree to which computer calculations can describe realistic systems. Also, in these cases, the theory is not nearly so well developed as in the simple fluid case. These mean that in the long term neutron measurements will be even more important in guiding us toward to an understanding of the properties of these systems.

Structural and dynamical properties of liquid surfaces, specially in water and electrolytes, have considerable interest from the point of view of a large number of phenomena of practical importance. Although experimental methods in this area are not yet developed we hope that more powerful sources and new techniques may stimulate work on liquid surfaces. It is possible for example that short wave-length neutrons can be used in total reflection techniques to study surface phenomena as a function of depth.

B. WAYS IN WHICH IPNS CAN BE USED FOR LIQUID RESEARCH

1. Experiments which make Use of the Special Properties of IPNS

There are two special features of an intense pulsed neutron source, which should be exploited for liquid state experiments. These are: (a) the pulsed nature of the system and (b) the intense $1/E$ spectrum produced by a small hydrogenous moderator.

It is worth emphasizing that a clean pulsed source allows experiments to be done in which the room background is switched off while the signal is being observed. This feature makes possible a number of experiments which are impracticable with steady state sources. While a high $1/E$ spectrum is not unique to a pulsed system, it is a natural feature which is more easily obtained than with steady state sources. A moderator should be optimized to give the maximum $1/E$ spectrum, so allowing high class scattering experiments to be done in the range of incident energies 0.1 - 100 eV. We discuss first, experiments which we can readily see taking advantage of these properties and secondly some speculative experiments which may utilize them in the future.

One outstanding problem in quantum liquids is the role of statistics in the determination of the macroscopic properties of the liquid. A direct demonstration of the role of statistics in this connection is to measure, by

means of inelastic neutron scattering at large momentum transfers, the fraction of atoms in the zero momentum state in superfluid ^4He . Several such experiments have been attempted in several laboratories but they were inconclusive and their interpretations model dependent. The reason for this is the upper limit of momentum transfer Q at which the experiment could be performed. This upper limit ($Q \lesssim 20 \text{ \AA}^{-1}$) was set by the low flux of neutrons at epithermal energies. Because of the nature of final state interactions, which are in turn determined by He-He scattering cross sections the value of Q at which it might be possible to observe a sharp peak arising from scattering by ^4He atoms in the condensate has been estimated to be $Q = 100 \text{ \AA}^{-1}$ or larger. Only by using incident neutrons of very high energy (10-100 eV) is it possible to extend in a significant way the range of Q values at which to search for and hopefully measure this extremely important parameter of quantum fluid physics.

A similar problem as far as technique of measurement is concerned is the determination of the mean square force on an atom in a liquid. This can be determined from measurements of the fourth moment of $S(Q, \omega)$ (see Appendix II) or, more simply, in the impulse approximation at high values of Q , the width of the Doppler broadened distributions which are centered on the recoil energy, $\omega = \hbar Q^2/2M$. Present day neutron sources do not have sufficient intensity at epithermal energies to be able to carry out such measurements in classical liquids to the accuracy required for meaningful theoretical interpretation.

There are some problems, such as the structure of glasses and of concentrated ionic solutions, for which data are required out to $50\text{--}100 \text{ \AA}^{-1}$. Such data allow naturally sharp radial distribution functions to be observed properly. Experiments with existing pulsed sources have shown that an intense $1/E$ spectrum can be used very effectively in this field, and we expect this method to be developed further.

The low room background between pulses may lead to a signal/noise ratio, compared to an equivalent steady state reactor experiment, about 10-100 times better. This advantage can be used in several ways, the likely ones being either to simplify the equipment or to observe weaker events. In the latter category are scattering experiments involving absorbing samples (e.g., the dynamics of liquid ^3He) or experiments where the process has a low cross section (e.g., high energy transfer events important to the determination of fourth moments of $S(Q, \omega)$). In the former category are experiments in which, e.g., a large number of detectors or analyzing crystals, are required. For both cases we may expect important liquid state experiments to be done which cannot be attempted now.

Because of the coupling between the energy and momentum transfer on scattering, a neutron experiment with a fixed incident energy cannot cover all the ω - Q ranges of theoretical interest. In particular most of the present experiments are deficient in the range where $Q < \text{\AA}^{-1}$ and $\omega > 10^{13}/\text{sec}$. This region is important in studying the growth of hydrodynamic modes from the assembly of microscopic modes in the intermediate, nonhydrodynamic regime, for example, in the onset of sound wave peaks as Q is reduced. It is also important in studying rotational behavior of molecular systems, and vibrational problems where Doppler broadening must be minimized. The use of small

angle inelastic scattering with high energy incident neutrons overcomes this problem, and the intense $1/E$ spectrum of IPNS will allow a large improvement over present sources.

2. Some Speculative Experiments

The above properties should enable new types of experiments to be done which would not be possible (or would be very difficult) on another source. The high flux combined with the pulsed nature of the source should enable the study of high intensity and of transient phenomena to be commenced. If the neutron burst is synchronized with a laser pulse (or a magnetic or pressure pulse) on the sample then $S(Q)$ or $S(Q, \omega)$ may be measured for large fields, or if the neutron pulse is delayed relative to the sample pulse then the variation of $S(Q)$ or $S(Q, \omega)$ with delay time may be measured. This type of experiment has excited interest in the past but has proved to be technically difficult. We believe that IPNS is likely to be sufficiently powerful, both in flux and for short-time scales, to make realistic experiments possible. High field experiments are likely to be the first application.

Another area which has given rise to speculation, but in which little work has been done, is the use of the wide range of wavelength available from a $1/E$ source to employ changeable scattering or absorption amplitudes. A simple case would be to reduce the effect of absorption in experiments involving He^3 or Cd samples. Another type is "anomalous scattering" near resonances to vary scattering amplitudes in diffraction experiments. Test experiments of this type have been carried out on steady state sources but the low flux in the 0.1 to 100 eV range has limited their usefulness. From the theoretical point of view these experiments are potentially valuable and the IPNS should be studied from this point of view.

The use of polarized neutrons to separate coherent and incoherent scattering is a proven method, but is of limited usefulness because of the low intensity and restricted wavelength range of polarized neutron beams. If wide band efficient polarizers become available, such experiments carried out on liquid sodium or water, for example, would be important.

Table I is a summary of the new proposals discussed in this section.

Table I. Summary of New Proposals

<u>Property of IPNS</u>	<u>Experimental Parameters and/or Conditions</u>	<u>Typical Experiments</u>
Low background	Low intensity and thus background limited	<ol style="list-style-type: none"> 1. $S(Q, \omega)$ for ^3He and other highly absorbing specimens 2. Fourth moment of $S(Q, \omega)$
High intensity of epithermal neutrons	Impulse approximation region of Q ($\gtrsim 15 \text{ \AA}^{-1}$)	<ol style="list-style-type: none"> 1. Condensate in ^4He 2. Mean square force on an atom
	Diffraction at high Q	Structure of glasses and ionic solutions
	Small Q , large ω	Transition from hydrodynamic to intermediate regimes ($Q \gtrsim 30 \text{ \AA}^{-1}$)
	Variable scattering or absorption amplitudes	<ol style="list-style-type: none"> 1. $S(Q, \omega)$ for specimens highly absorbing in the thermal region 2. Partial diffraction patterns
Pulsed nature of source	Time dependent experiments	Study of high field effects or of transient phenomena following impressed laser, pressure or magnetic field pulses

3. Comparison with CP-5

At present there is a viable liquid state program on CP-5 and there are many interesting liquid state problems which may be studied with the wavelength range covered by thermal reactors. It is important that IPNS be able to cover such experiments as well as the new areas listed above. The flexibility of the moderators on a pulsed source should enable moderators to be tuned for 1/E, thermal or cold neutron regions. This factor, together with the high flux will enable the present range of experiments to be improved significantly compared to the existing facilities at CP-5. We regard both this improvement and the coverage of existing ranges as important factors.

C. Equilibrium Properties

The static structure factor, $S(Q)$, is an important piece of information for the purpose of obtaining interparticle potentials specially in monatomic liquids; moreover, it is necessary to know $S(Q)$ at various temperatures and densities. Up until now this important property has been investigated with requisite accuracy for only a very few liquids and at isolated points in phase space. Liquid argon for instance has been measured with high accuracy only at its triple point. Developments in the theory of the liquid state on the one hand and computer simulation methods on the other require diffraction data of high accuracy on a fairly dense grid of temperature and density; the former will make it possible to derive a state dependent pair potential directly from experiment while the latter will be able to obtain a potential consistent with the data. A thorough determination of a state dependent pair potential will make it possible to determine the 3-body contribution to the interaction potential in the system. This approach is of particular importance in the study of metals where many-body contributions can be expected to be considerable.

In the case of ionic systems like molten salts the pair correlation $g_{+-}(r)$ depends sensitively on the ionic interactions; it is only recently that the effect of the polarizability of ions on g_{+-} has started to be investigated; otherwise the rigid ion picture of the ions was assumed to be valid for ionic melts. Neutron diffraction studies using isotopic substitution has been employed for obtaining partial structure factors S_{++} , S_{--} , S_{+-} , and hence the pair correlation g_{+-} . Here again one has to emphasize the necessity of high accuracy data because the method depends on the subtraction of two pieces of data.

It is possible to vary the coherent scattering from water by an appropriate mixture of H_2O and D_2O . This method needs to be further developed as a means of studying the partial pair correlation function, which in turn will give us a better understanding of the structure of water specially under extreme conditions of temperature and pressure.

In the theory of monatomic liquids the three body correlations are only partially understood. It is known that a determination of the density derivation of the structure factor makes it possible to test the theory of the triplet correlation function. This example again hinges on the accuracy with which the equilibrium structure can be determined experimentally.

The $1/E$ spectrum of the proposed pulsed source will be a special tool and will make it possible to obtain high resolution structural information through diffraction measurements out to $50\text{--}100\text{ \AA}^{-1}$. Some applications of this capability are the following.

The network structure in glasses such as Ge, Si, As, C, etc., cannot be properly understood without a reliable determination of $g(r)$, the pair correlation function, in the neighborhood of each atom. For this purpose it is necessary to measure $S(Q)$ to values of Q up to 50 \AA^{-1} or larger. Such measurements are needed for meaningful Fourier inversions to obtain sharply defined $g(r)$ in the first few coordination shells.

For glasses containing more than one atomic species the data cannot readily be interpreted, as is the case of monatomic glasses. However isotopic substitution could again be of great importance; the proposed facility will enable us to get information of the necessary accuracy, in the high Q range, without which the Fourier inversions cannot be made. Chalcogenide glasses and semiconductors (As, Se, Ge, Te compounds) have considerable technological importance; their structure needs to be well understood if one attempts a clarification of their electronic properties.

Another area is that of the structure of concentrated ionic solutions, where it is believed that the ionic order may be longer ranged than for a simple liquid, and the ions are localized for long times. In such cases measurements to high Q are needed to establish the structure.

The time-average structure factor $S(Q, \omega = 0)$, is of some theoretical interest in liquids and glasses. For these cases where the structure is long-lived this function may be interpreted in a straightforward manner, but for short-lived structures further theoretical study is needed. It is possible that by the time that IPNS is built, measurements of this type will be more widely used than at present. For this reason we shall discuss a spectrometer for such measurements in Section F.

Time-of-flight spectrometers of existing designs seem to be adequate for diffraction experiments. It would appear that the reliability of the desired results is at present limited by the accuracy of the corrections which have to be applied to the data (multiple scattering, dynamic effects). These corrections are less well understood in the case of pulsed than of steady state sources.

D. Dynamical Phenomena

1. Gases

One important problem in fluid state calculations is in treating the competition between simple binary collisions, which dominate at low densities, and collective mode-coupling effects which become increasingly important at high densities. These effects most strongly influence the transition region between hydrodynamic and kinetic behavior. This transition region has been investigated at liquid densities using both neutrons and computer molecular dynamics, but the neutron data are of limited accuracy. Light scattering has been used to investigate this transition in the low density region, but this

technique is not useful for more dense systems. The intermediate density region has not yet been investigated by any technique. It is highly desirable to have good neutron and computer data in this region since we can, as we increase the density, turn on the interesting collective recollision effects. For this reason, careful and controlled high resolution experiments on a monatomic gas, like Ar^{36} , should be performed to provide experimental data in a system dilute enough to be described in terms of binary collisions. By extending such measurements to higher densities the onset of higher order collective effects can be carefully monitored.

Density dependent studies offer intriguing possibilities of raising the level of our understanding of binary systems and of complex molecular fluids such as water. Two interesting binary gas systems in which the second component is either heavier or lighter than the host gas are the $\text{Ar}^{36}\text{-He}$ and $\text{Ar}^{36}\text{-Xe}$ systems. In these systems, the scattering is almost entirely dominated by Ar^{36} and the He or Xe components are largely unobserved impurities which modify the $\text{Ar}^{36}\text{-Ar}^{36}$ density fluctuations. The density dependent $S(Q, \omega)$ should be describable in terms of the generalized Enskog equation and it is of great interest to fundamental statistical theories to test the validity of this equation at intermediate densities. By studying simple systems at a wide variety of temperatures and densities it should be possible to provide the data base necessary for extending our knowledge of many-body effects which ultimately govern all fluid behavior. Such studies should provide the basis for extending our knowledge of simple fluids to the more complex fluids such as N_2 , Br_2 , CO_2 and H_2O .

2. Dynamics of Dense Liquids

a. Monatomic Systems

Accurate measurements have recently been performed in the intermediate wavevector and energy range ($1 \text{ \AA}^{-1} < Q < 5 \text{ \AA}^{-1}$ and $0 < \hbar\omega < 10 \text{ meV}$) for several classical liquids, and the data have been shown to be precise enough to satisfy the zeroth and second moments of $S(Q, \omega)$. These results have provided a valuable testground for theoretical calculations. In particular, the fourth moment of $S(Q, \omega)$ can be used to give a determination of an effective pair potential, $V(r)$ complementary to that derived from neutron diffraction. A comparison of these two effective potentials should be useful in understanding the role of many-body forces.

In liquid metals, where pseudopotentials are in common use, such a method would be of great interest as input for microscopic theories. The feasibility of measuring the fourth moment has been investigated briefly and a report is given in Appendix II. The measurements of $S(Q, \omega)$ to desired accuracy would be rather time consuming at presently available facilities, limiting the studies to very few thermodynamic states. IPNS, with its increased flux and especially the low background, would provide a possibility for measuring $S(Q, \omega)$ for a variety of pressures, temperatures, and densities, yielding more extensive experimental data than presently available. This might in turn provide a key to a more fundamental understanding of the liquid state.

At relatively small wavevectors ($Q \lesssim 1 \text{ \AA}^{-1}$) the measurement of collective excitations is difficult experimentally. In this region where ordinary hydrodynamic theory does not apply, the dynamical behavior of liquids as simulated on computers shows collective modes, the nature of which is not understood. They do not obey simple hydrodynamics, but appear to have some characteristics reminiscent of zero sound propagation, so far only observed experimentally in crystalline solids and liquid helium. In these experiments one has not yet obtained the accuracy needed for a detailed comparison with the calculations; but the experience gained seems to indicate that a mere increase of neutron flux would improve the experimental accuracy to a satisfactory level.

The large wave vector region ($5 \text{ \AA}^{-1} < Q < 20 \text{ \AA}^{-1}$) where even in the dense liquid the atomic motions are largely uncorrelated, kinetic theories have explained the measured neutron spectra at a semi-quantitative level. The correlations are described by a mean force exerted on the atoms, but a better insight would be gained if one could probe the excitations at higher Q into the weakly correlated regime and the transition to the uncorrelated free particle-regime. This requires, according to present theories, extremely large wavevector and energy transfers.

The special attention devoted to superfluid ^4He has made this the most extensively studied liquid. The neutron scattering results have provided a stringent test of theoretical ideas, including direct evidence that the Landau hypothesis of the existence of well-defined excitations (the well known "phonons" and "rotons") is essentially correct. We expect this interest to continue with similar detailed measurements in the nonsuperfluid phase and at elevated temperatures and pressures. In particular, as proposed above, large wavevector experiments in liquid ^4He might provide crucial insight into the existence of the superfluid condensate.

b. Molecular Liquids

These liquids have complicated dynamical properties. Both rotational and translational motions contribute to the neutron spectra, and it is of interest to study the effect of the coupling between the two types of motions. Experiments have not so far provided a very detailed insight into this effect, and a breakthrough in the study of molecular liquids is not immediately foreseeable.

The dynamics of liquids composed of simple linear molecules is currently being studied in a number of laboratories. For more complicated molecules including liquid crystals, for molecular glasses and for ionic solutions inelastic neutron scattering data are difficult to interpret partly because the spectra are complex and partly because the structure of these systems is poorly understood. It is, however, quite likely in view of the progress made over the past few years in simple liquids, that by the time IPNS is available our understanding will have improved enough to place these important classes of liquids into the center of interest.

The special case of glasses has received some attention in the literature and the properties of fundamental modes of motion have been considered. To improve this field would require similar instrumentation to that suggested for liquids, probably with higher resolution at low Q .

c. Molten Salts

Alkali halide crystals are classic materials in the field of Solid State Physics and the study of their structure, dispersion curves and thermodynamic properties have a long history. It is only recently that microscopic study of the structure and dynamics of molten salts has been undertaken. Due to the long range nature of the Coulomb interaction, charge neutrality plays a dominant role in the structure and the dynamics of these materials. However, the order, apparently minor, details of the interaction potential actually determine the properties of specific molten salts. For example in molten NaCl the coordination number of first neighbors is 6 whereas in BeF_2 there are four F's around each Be; and the latter has a viscosity several orders of magnitude more than that of the former. It is well known that LiF, BeF_2 mixtures have a lower viscosity than BeF_2 by itself. Our understanding of these properties in terms of ionic sizes is poor and the detailed nature of microscopic dynamics that causes these phenomena is not understood. It is therefore appropriate to study these materials with neutron inelastic scattering to determine the various partial dynamic scattering factors. By an appropriate choice of ionic sizes, charges and nuclear scattering amplitudes, neutron inelastic scattering can bring into focus different aspects of the dynamics of particles in these systems.

3. Diffusion Studies

Incoherent neutron scattering is a useful method for observing single particle motion in dense fluids on an atomic scale. Experiments have given tests for models of the single particle motion (especially the nature of the diffusive step) in liquids and gases; and measured diffusion constants mostly agree with other experimental results, e.g., with NMR. Neutron scattering is, however, the only experimental method of measuring diffusive motion in the finite Q and ω regime, and it now appears possible to test fundamental microscopic theories derived from kinetic equations by measuring the density dependence of incoherent scattering from gases at dilute, intermediate and high densities.

Substantial interest has been devoted to incoherent neutron scattering from homogeneous molecular liquids. Such experiments yield information about both translational and rotational relaxations, and a fair agreement with NMR, IR, and Raman scattering has been obtained. Systematic studies have illuminated the molecular shape dependence of diffusive effects. There are a number of models to describe this behavior, and careful high resolution measurements are necessary in order to improve these models.

A problem which in its technological aspect is related to the studies described in Sec. E is the case of the diffusion of the isotopes of hydrogen in molten lithium. Molten lithium may be used as blanket material in the CTR-reactor and for this purpose, the transport coefficients of the hydrogen isotopes must be known. It has not been possible so far to obtain the diffusion constant from macroscopic measurements whereas in the case of neutron scattering this can be done under well controlled conditions.

E. LIQUID-LIKE DIFFUSION IN SOLIDS

The physical properties of the transition metal hydrides have been studied extensively over a period of time. The last few years have seen an increase in the efforts in this field. This is due to the realization that several of these systems are of potential interest for a number of technological applications. For example, Th_4H_{15} and PdH_x have been found to be high T_c superconductors and several of the hydrides are considered for the storage of hydrogen as part of a hydrogen based energy scheme. Also vanadium and niobium are the most likely candidates for first wall material in future CTR-reactors. In both of these applications it is necessary that the transport properties of hydrogen be well understood. The quasielastic neutron scattering method is capable of measuring the details of the diffusion process on an atomic scale. As a result of these studies we have reached a good understanding of the diffusion process in fcc metals (PdH_x) at low concentrations of hydrogen. However, the results obtained so far for bcc systems (V, W, Ta) show significant deviations from any proposed model for the jump process. Future progress in this area requires a refinement of the theoretical description of the diffusion process and the accumulation of neutron scattering results of higher precision.

Another case of technological interest is atomic transport in solid electrolytes, e.g., β -alumina. In this case the interest arises from the possible use of these materials as components in batteries; the understanding of the ionic transport is of course crucial to this application. The neutron method is again capable, in principle, of giving the detailed information that is needed for a basic understanding of the phenomena. These systems should be studied at room-temperature, which is the ambient temperature in the battery application, and for this purpose measurements at higher resolution than can be obtained at present facilities will be required.

The rotational diffusion in plastic crystals is similar to the same process in liquids. This area is of interest to liquid state and molecular crystal physics, but present work is limited by the available spectrometers. We hope that improved sources and spectrometers, especially low Q spectroscopy will lead to a better understanding.

The diverse research areas brought together in this section pose similar requirements on the experimental equipment, as do the fields discussed in Sec. D.

F. COMMENTS ON INSTRUMENTS

We have considered three instrument proposals:

- (i) large energy transfer small momentum transfer spectrometer
- (ii) general purpose time-of-flight spectrometer
- (iii) high resolution time-of-flight diffractometer

It is probably that these three instruments would be used for the program of measurements discussed in this report, but that modification of them

or new instruments would be needed to cover all of the proposed experiments. The major changes would be:

The range of machine(s) is insufficient: in addition a special small angle inelastic scattering spectrometer should be built along the lines illustrated at Fig. 1. Wavevector range is 0.05 \AA^{-1} to 1 \AA^{-1} and energy transfers dependent on the incident energy and the velocity of sound in the specimen (assumed to be 1000-6000 m/sec). 0.05 to 1.0 eV neutrons are required to satisfy both momentum and energy conservation. The scattering angles should be in the range 0.1° to 2° with resolution of $\pm 0.05^\circ$.

The general purpose time-of-flight spectrometer does not have the range to cover the impulse-type experiments: for such work a resonance-detector down scattering spectrometer is required as illustrated in Fig. 2. For a flight path of 10 m, a sample area of 100 cm^2 and a resolution of 30 meV, the counting rates at 6.7 eV are large enough to measure a spectrum in a short time. Several angles of scatter should be studied simultaneously and several resonance absorbers should be used (for energies from 1 to 100 eV). For some of these experiments short neutron pulses of $\sim 0.1 \text{ \mu sec}$ are required.

For liquid diffraction work an instrument of the type (iii) is required but the resolution and range should be adjusted to liquid diffraction patterns (0.1 to 100 \AA^{-1} with $\Delta Q/Q$ varying from 0.2 to 0.01): this may require a special machine if the powder diffraction instrument is heavily used. A similar range is required for elastic diffraction, but we have not considered the machines that would be needed for the whole range. A machine is suggested below (d) for $Q < 10 \text{ \AA}^{-1}$.

An $S(Q,0)$ elastic scattering diffractometer would analyze a diffracted beam in both initial and final energy, detecting only those neutrons that have been elastically scattered. Inelastically scattered neutrons have a wavelength that does not correspond to the wavelength for Bragg reflection. Elastically scattered neutrons satisfy the Bragg condition of the rotating crystal at the proper time. The rotating crystal must move at an angular velocity such that at each instant

$$\theta = \sin^{-1} \left(\frac{ht}{2mLd} \right). \quad [\text{see Fig. 3}]$$

We also note that in some cases provision could be made for two instruments to be installed on one beam hole, and therefore a large experimental area will be required. In addition, if the emphasis on the $1/E$ region is confirmed, then many experiments using long flight paths ($\sim 20 \text{ m}$) are likely to be required. Finally some instruments, e.g., the position sensitive time-of-flight spectrometer and the resonance γ -ray time-of-flight detector, are multiparameter detecting systems and sufficiently elaborate data handling and recording systems will have to be provided.

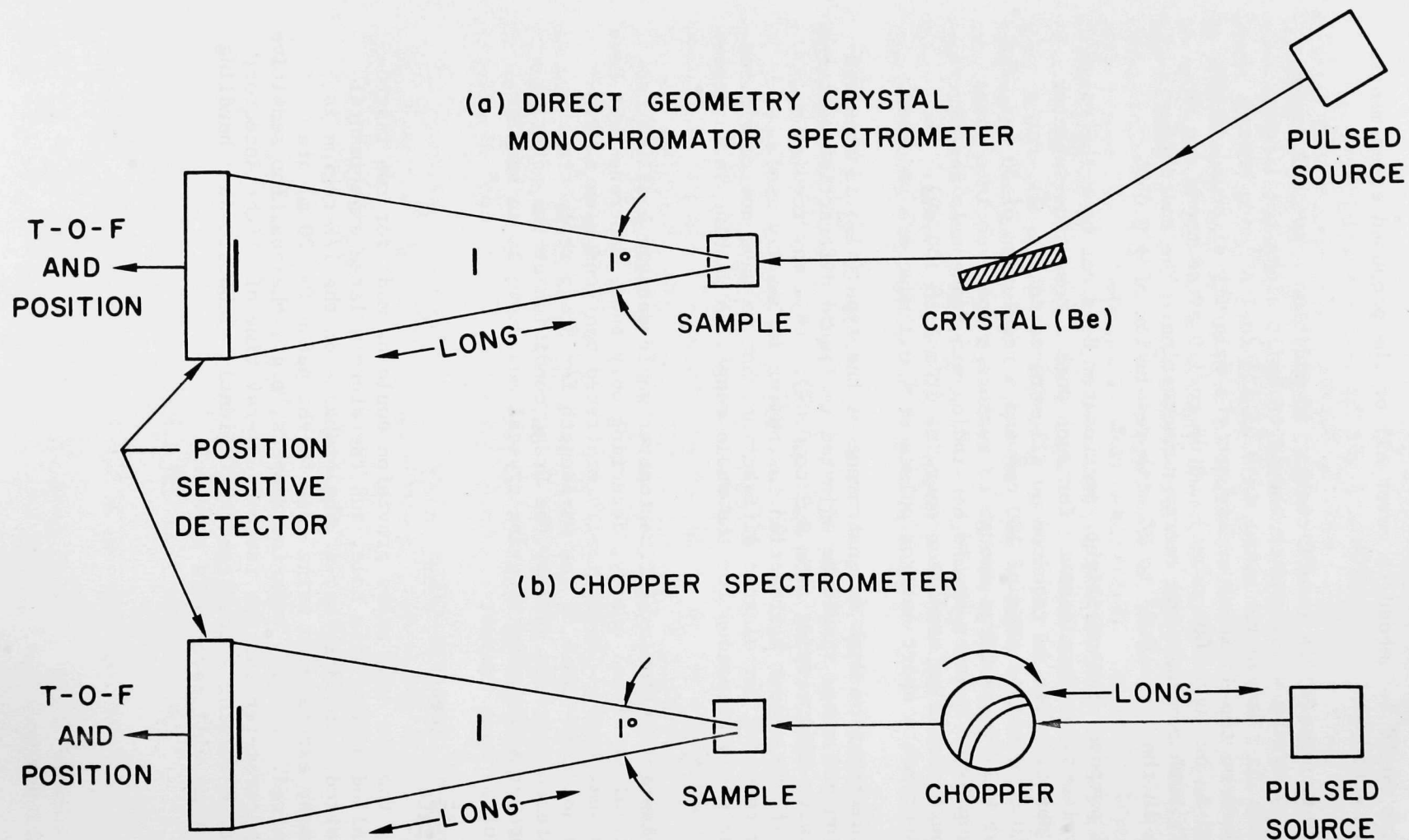


FIGURE 1

Small angle inelastic scattering spectrometers

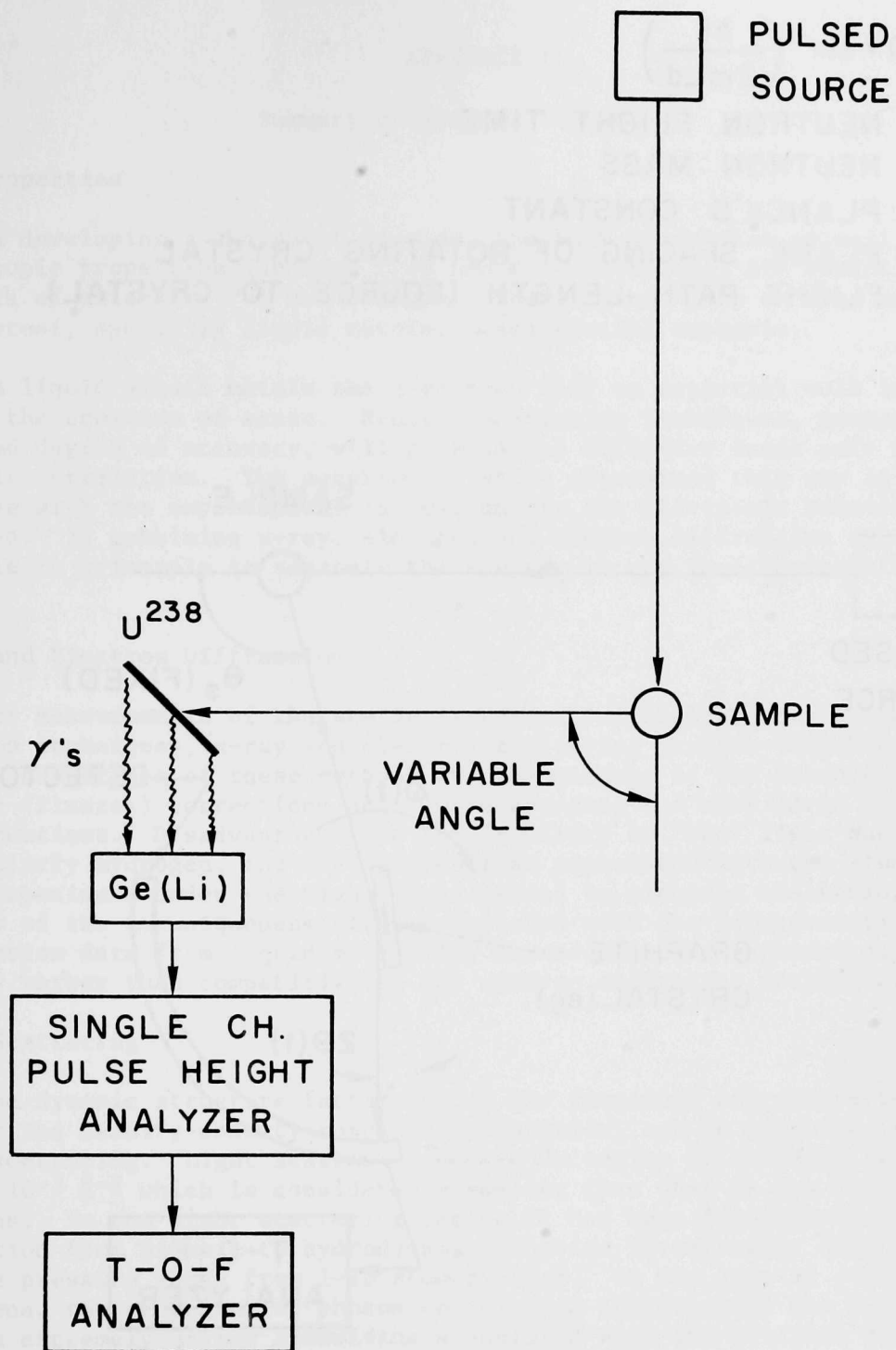


FIGURE 2

RESONANCE DETECTOR SPECTROMETER

$$\theta(t) = \sin^{-1} \left(\frac{ht}{2mLd} \right)$$

t = NEUTRON FLIGHT TIME

m = NEUTRON MASS

h = PLANCK'S CONSTANT

d = PLANE SPACING OF ROTATING CRYSTAL

L = FLIGHT PATH LENGTH (SOURCE TO CRYSTAL)

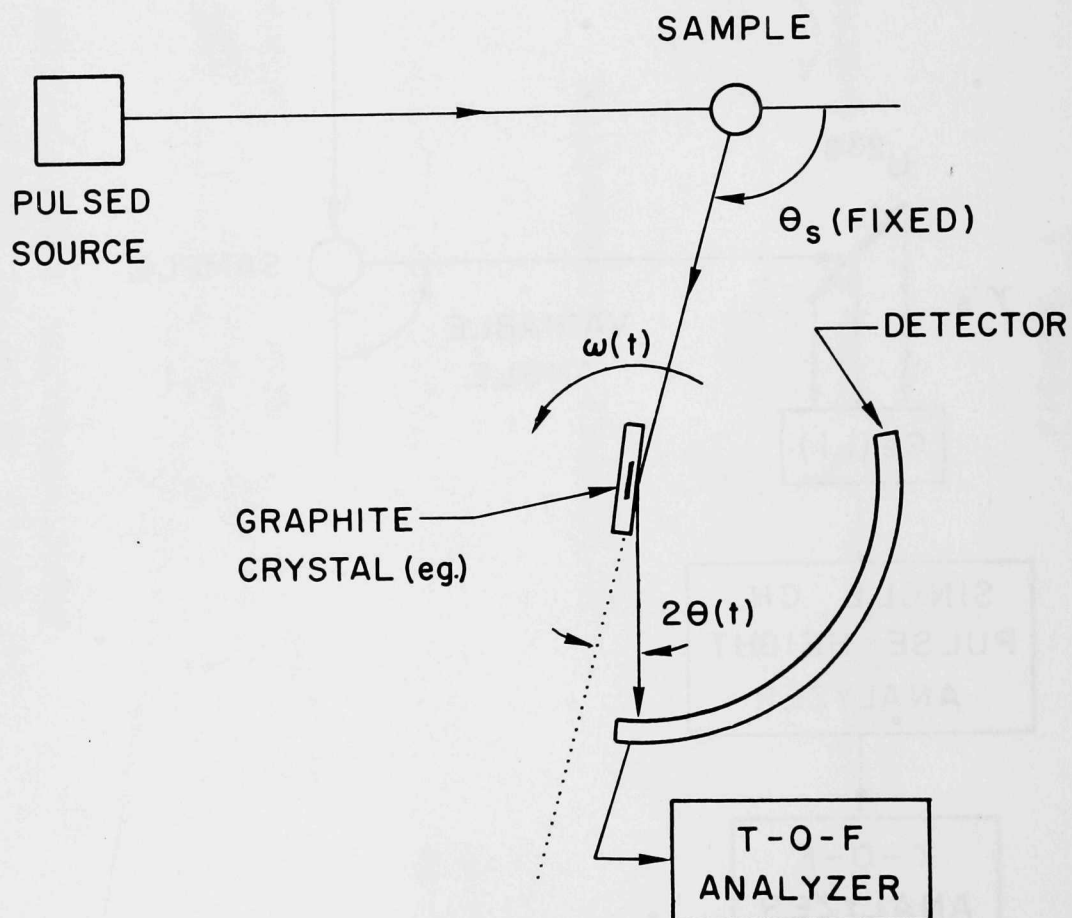


FIGURE 3

S(K,0) ELASTIC SCATTERING DIFFRACTOMETER

APPENDIX I

Summary of Several Techniques

Bulk Properties

In developing a theory of liquids, the bulk properties as well as the microscopic properties (deduced from neutron scattering) are essential. The lack of extensive PVT and transport coefficients data on many materials of interest, specially liquid metals, needs special emphasis.

In liquid alkali metals the electrons play an essential role in determining the equation of state. Neutron scattering techniques, pushed to the required degree of accuracy, will give us the effective ionic pair potential and pair correlation. The equation of state determined this way is bound to disagree with the experimental values, unless the electronic behavior is included. By combining x-ray, electron and neutron diffraction data it is possible in principle to separate the electronic and ionic correlation functions.

X-ray and Electron Diffraction

For measurements of the static structure function by conventional diffraction techniques, x-ray and electron scattering provide valuable alternatives. Advantages of these methods are the absence of the troublesome dynamic (Placzek) corrections and, for electrons, the very large scattering cross sections. Disadvantages are the inability to "see" light nuclides, particularly hydrogen, and the difficulties associated with the study of liquid specimens under the high vacuum needed in electron scattering studies. In view of the nonuniqueness often associated with the interpretation of diffraction data from liquid materials, these should be regarded as complementary rather than competitive to the neutron scattering technique.

Light Scattering

The dynamic structure factor $S(Q, \omega)$ the frequency and wavevector transform of the density-density correlation function, can be measured in part by light scattering. Light scattering senses the region of momentum transfers around 10^{-3} \AA^{-1} which is considerably smaller than what is now available with neutrons. In the light scattering regime it has been possible to observe the transition from kinetic to hydrodynamic behavior in extremely dilute gases (in the pressure range from 1-10 atmospheres). In the area of critical phenomena, the technique of photon correlation spectroscopy has been used to measure extremely narrow linewidths associated with the critical slowing down of entropy fluctuations due to mode coupling effects.

Depolarized light scattering has been used extensively to study molecular reorientation phenomena and dimer state formation in molecular and simple liquids. This field has also produced significant measurements with anisotropic processes in liquids.

While light scattering will continue to make contributions to our understanding of molecular rotations and collision phenomena in liquids, it is in

many ways a macroscopic probe. Also there are many liquids for which light scattering is unsuitable because of the attenuation of light by the samples.

NMR Experiments

For a number of liquids (especially homogeneous ones), this technique has proven useful in measuring macroscopic diffusion constants and rotational motion to a degree of accuracy which at present exceeds that of neutron scattering. It usually gives information outside the ranges covered by neutron scattering and in many cases its very high selectivity is an important asset. The interpretation of data often depend on models, so that fundamental neutron scattering work can assist the NMR field.

Infra-red Absorption and Raman Scattering

Molecular energy levels and reorientational motion is at present more accurately studied by IR and Raman scattering, than in neutron scattering where unavoidable Doppler broadening smears out the molecular spectra. High energy neutrons might improve this situation and make the techniques complement one another. Then one could make use of the fact that absolute neutron scattering cross sections are more easily calculated than in many optical experiments; and also the lack of selection rules might become of more quantitative use (see Chemical Spectroscopy report).

APPENDIX II

Fourth Frequency Moment of the Scattering Function

It has been demonstrated recently that a straightforward computational scheme allows the extraction of the effective pair potential from the fourth frequency moment ($\langle \omega^4 \rangle$) of the scattering function. Application of the method to a real case has not been possible because sufficiently accurate results for the scattering function have not been obtained so far for any liquid.

The major experimental difficulty is that the scattering function must be determined accurately for large values of ω over a wide range of Q . This poses two specific problems: (i) the intensity in the experimentally observed function is very low for large values of ω and the signal is lost in the background noise, and (ii) the measurement at large ω and small Q is kinematically possible only if neutrons of high energy are used. Therefore the spectra obtained using present reactors are not suitable in this respect. However, two special features of the IPNS source are that the background is low between the source pulses and that the spectrum is rich in epithermal neutrons.

In order to estimate whether such an experiment is indeed feasible at IPNS, we have made a Monte Carlo simulation of a "thought" experiment in which $\langle \omega^4 \rangle$ is determined for liquid argon in the range $1 < Q < 4 \text{ \AA}^{-1}$. The assumption is made that the scattering function can be represented by a gaussian memory function for the current-current correlation function with $\langle \omega^4 \rangle$ determined from a L-J potential and recent accurate experimental results for $g(r)$. This model is in good agreement with available results for the scattering function for liquid argon. The experimental parameters are those predicted for the proposed IPNS facility. The "observed" intensity is generated by Monte Carlo simulation and standard data reduction technique is used to extract the scattering function and the fourth frequency moment. By this simulation we verify that one should obtain the $\langle \omega^4 \rangle$ moment within an accuracy of 5% from a real experiment. This encourages us to believe that it will be possible to measure $\langle \omega^4 \rangle$ with enough precision that useful information about the effective pair potential can be obtained.

VII. REPORT OF THE PANEL ON MAGNETISM AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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MAGNETISM

A. GENERAL INTRODUCTION

As a result of the rapid fall-off of the magnetic form factor $f_m(\vec{q})$ with increasing \vec{q} for most materials of interest in magnetism, the range of scattering vectors of interest is usually $Q \lesssim 4 \text{ \AA}^{-1}$. Thus, in order to complete the scattering triangle, satisfying conservation of both momentum and energy, one needs a rather high incident neutron energy E_0 to observe excitations of rather modest energy $\hbar\omega$. Consequently, the large number of neutrons coming from a fast-pulsed neutron source in the energy range of 0.1 - 2 eV are extremely important. In Fig. 1, for a wavevector transfer $Q = 4 \text{ \AA}^{-1}$, we plot the minimum incident energy E_0 necessary to observe an excitation of energy $\hbar\omega$. The corresponding scattering angles are also shown.

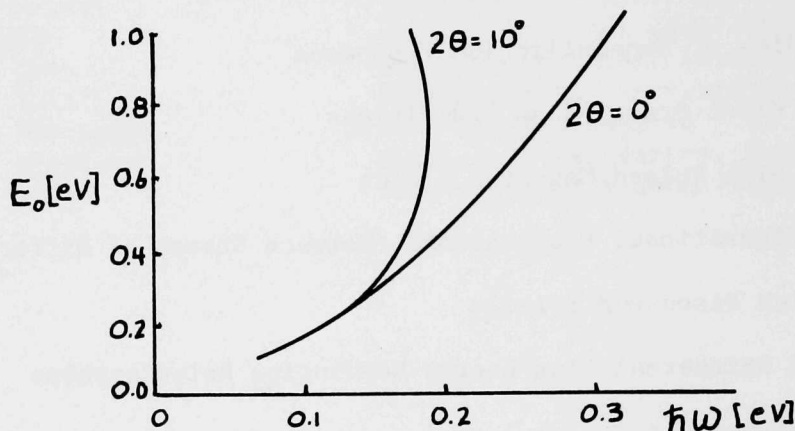


Fig. 1: Minimum incident neutron energy, E_0 to observe an excitation of energy transfer $\hbar\omega$ with $Q = 4 \text{ \AA}^{-1}$.

The scientific motivation for wanting to build a source such as IPNS originates, in part, from previous unsatisfactory experiments attempting to extend our understanding of magnetism to a more and more microscopic level. An example of these are the excitations in the 3d transition metal ferromagnets Fe and Ni--or more precisely, the measurement of the generalized susceptibility $\chi(\vec{q}, \omega)$. Another example is the various attempts to measure the spin wave spectrum in the spin density wave (SDW) antiferromagnet, Cr metal. Also, there are many cases where inelastic polarization experiments have been desirable to obtain directly information on the relative magnitudes of nuclear and magnetic scattering, the parallel and perpendicular parts of $\chi(\vec{q}, \omega)$, and also the separation of spin incoherent and coherent scattering. Experiments of this type become feasible with the projected incident beam intensities available at IPNS. In fact, even at $\lambda_0 = 1 \text{ \AA}$ ($E_0 \approx 80 \text{ meV}$) a comparison of the flux from IPNS with the high flux reactor HFIR at Oak Ridge is impressive. Roughly, the nominally monochromatic intensity ($\Delta E/E \approx 0.02$) available at a sample for inelastic experiments, using a single crystal analyzer, at HFIR is $2 \times 10^7 \text{ n}\cdot\text{cm}^{-2}\text{s}^{-1}$. At IPNS the projected time-averaged incident flux available at the sample (7 m from the moderator) for inelastic measurements, by time-of-flight analysis, is $10^9 \text{ n}\cdot\text{cm}^{-2}\text{s}^{-1} \text{ eV}^{-1}$. Although all experiments cannot use the entire energy spectrum in this incident beam, a large number of experiments can use a substantial fraction of these neutrons.

For example, if we have an experiment, for which the cross-section (or other requirements) limits us to the band of energies 70 meV to 110 meV, then the "effective time average flux" at the sample is $(0.04 \text{ eV}) \times (10^9 \text{ n}\cdot\text{cm}^{-2}\text{s}^{-1} \text{ eV}^{-1}) = 4 \times 10^7 \text{ n}\cdot\text{cm}^{-2}\text{s}^{-1}$. However, if the band of neutrons which we can effectively use extends up to say 1 eV from some lower limit of, say 70 meV, as is the case in measuring $\chi(\vec{q}, \omega)$, for example, in the transition metals Fe, Ni, Cr, then the "effective time average flux" is $(1-0.07) \text{ eV} \times 10^9 \text{ n}\cdot\text{cm}^{-2} \text{ s}^{-1} \text{ eV}^{-1} = 93 \times 10^7 \text{ n}\cdot\text{cm}^{-2} \text{ s}^{-1}$. Thus, for this type of experiment, IPNS would be about 50 times better than a high flux reactor (see Sec. H for other details).

The pulsed nature of the source allows us to explore new regimes of applied magnetic field, perhaps up to 500 kG. Transient phenomena using pulsed fields, and high pressures, or both simultaneously, are well-suited for IPNS.

The panel treated some topics in detail. while others in only a cursory manner. The selection was dictated by the desire to choose examples where it was possible to evaluate quantitatively the performance of IPNS in problems where the use of existing high flux reactors have proved inadequate or impossible. A list of topics discussed briefly is given in Sec. G. The topics treated in some detail were:

- Applications of polarized neutrons
- Constant \vec{Q} time-of-flight spectrometer with its possible application to the Cr metal problem.
- Generalized susceptibility measurements in the transition metals.
- Transient phenomena near first order-phase transitions.
- Configurational fluctuations.

B. POLARIZED NEUTRONS

The interaction between the neutron and electron spin provides a unique tool in the investigation of magnetic phenomena. To utilize this interaction we require a method of differentiating between the two neutron spin states, either by producing a beam of polarized neutrons or analyzing the spin state of the scattered beam. In general, a knowledge of the initial and final neutron spin state always provides more information about the system. In many experiments the loss of flux associated with such an analysis is too great to tolerate. However, we anticipate polarized-beam experiments to become more important as the experiments become more sophisticated, probing greater microscopic detail of magnetic materials. At this time we are able to identify a number of areas which would benefit from using polarized neutrons. In particular the separation of magnons and phonons, the characterization of time-dependent magnetization effects (preferably in conjunction with a pulsed field), studies of spin glasses, amorphous magnets, diamagnetism, nuclear magnetism, magnetization densities, and general magnetic phase diagram investigations all benefit from the use of polarized neutrons.

Spin discrimination in a beam of white neutrons is a problem we have considered in some detail. In addition, we present preliminary considerations of an instrument, together with intensity estimates.

1. Polarizing Methods

Thermal and epithermal neutrons may be polarized by: (a) Bragg scattering from ferromagnetic single crystals, e.g., FeCo or Heusler Alloy; (b) spin-dependent scattering from polarized nuclei in a filter, e.g., a polarized proton target; (c) spin-dependent absorption in a filter, e.g., from polarized ^{149}Sm or ^3He nuclei; (d) using the Stern-Gerlach technique; or (e) total reflection from magnetized mirrors, e.g., as in a guide constructed of Permendur (Co(48), Fe(50), V(2)) mirrors. Of these methods the only techniques suitable for efficiently polarizing a white beam are (b) and (e). We have estimated the overall transmittances of two polarizers of these types using the following parameters.

Polarizing Guide: Magnetized Permendur Cobalt (50):Iron (50),
(Critical glancing angle/wavelength = $0.1^\circ \text{ \AA}^{-1}$).

Polarized Proton Filter: Proton polarization $\sim 90\%$
Filter thickness = 2 cm for a lanthanum
magnesium nitrate polarized target.

The results of these calculations are shown below:

TABLE 1. Performance of Polarizing Devices

Neutron Energy (meV)	Transmittances (%)		Polarizing Efficiency of Filter
	Guide	Filter	
327	2	21.0	0.898
82	4	23.2	0.960
5	16	32.3	0.998

We estimate the polarizing efficiency of the guide to be ~ 0.95 . Apart from the undesirable energy-dependence of the transmittance of the guide, it should be noted that its performance is not matched to the most favorable feature of the energy spectrum of the IPNS source; i.e., the intensity gains it provides at the higher neutron energies. A further disadvantage of the polarizing guide is that we expect its effective area (~ 1 or 2 cm^2) to be considerably less than should be achievable with the filter (~ 10 or 20 cm^2). Although the polarizing guides might be a factor of 10 cheaper to construct, we consider the polarized proton filter to be the most suitable polarizer for the white beam.

In order to spin-flip the polarized beam we recommend the use of a non-adiabatic spin-flipper such as a Dabbs foil (current sheet) or a two-coil "Drabkis" flipper; both these flippers have high efficiencies ($\sim 99\%$) which are essentially velocity independent, and are ideal for fast neutrons.

2. Proposed Spectrometers: Intensity Estimates

The direct approach to measuring polarization dependent cross sections is based on the fact that such a scattering process will result in polarization creation. That is, the scattered beam will be partially polarized when the incident beam is unpolarized. This effect can be measured with a polarizing crystal after the sample

It may be desirable to build a spectrometer along the lines illustrated in Fig. 2.

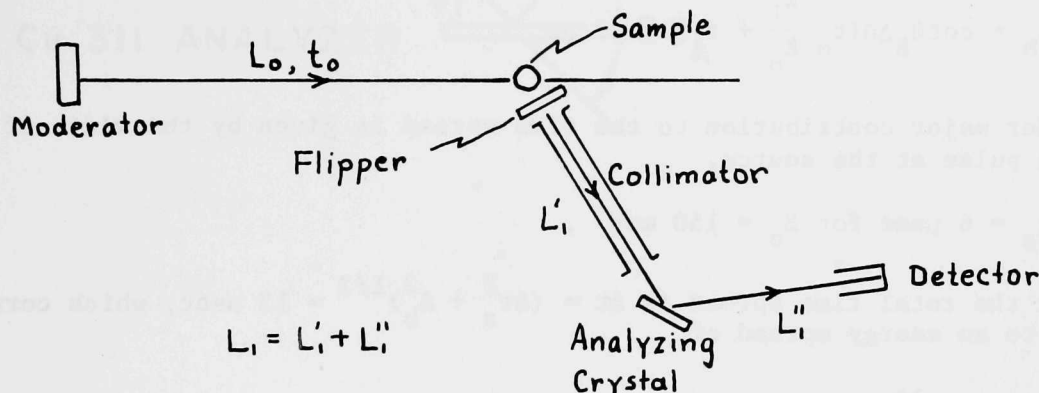


Fig. 2. A time-of-flight polarized neutron spectrometer.

The analyzer acts to select a final energy and final spin state. The combination of flipper and analyzer allows the selection of either final spin state. For an initial unpolarized beam the cross sections observed are

$$\frac{d^2(\sigma_{\uparrow\uparrow} + \sigma_{\downarrow\uparrow})}{d\Omega dE} \text{ with the flipper off and } \frac{d^2(\sigma_{\downarrow\downarrow} + \sigma_{\uparrow\downarrow})}{d\Omega dE} \text{ with the flipper on.}$$

With the final energy fixed by the analyzer crystal the time-of-flight analysis enables the determination of the initial neutron energy and energy transfer.

Addition of a polarized proton filter in the incident flight path would convert this instrument into a polarization analysis instrument capable of measuring both spin-flip and non-spin-flip inelastic cross sections.

With such a spectrometer it is desirable to keep the scattered path length small compared to the incident path length because the incident energy is determined by subtracting the final flight time from the total flight time.

For illustration, we select $L_0 = 9$ meters and $L_1 = 1$ meter. For a scattering process in which the neutron energy changes from 150 meV to 50 meV, and assuming 0.2 collimation before the analyzer and 0.2° mosaic spread in the analyzer, the following numbers are obtained:

$$t_o = 1493 \text{ } \mu\text{sec}$$

$$t_1 = 323 \text{ } \mu\text{sec}$$

$$\theta_B = 18.6 \text{ for } d = 2.0$$

$$\Delta E_1 = 2 E_1 \theta \cot \theta_B = 1.5 \text{ meV} \quad . \quad (1)$$

This energy spread results in a contribution to the total time spread given by

$$\Delta t_b = \cot \theta_B \Delta \theta (t_o \frac{E_1}{E_o} + t_1) = 12 \text{ } \mu\text{s} \quad . \quad (2)$$

The other major contribution to the time spread is given by the width of the neutron pulse at the source,

$$t_a = 6 \text{ } \mu\text{sec for } E_o = 150 \text{ meV} \quad , \quad (3)$$

so that the total time spread is $\Delta t = (\Delta t_a^2 + \Delta t_b^2)^{1/2} = 13 \text{ } \mu\text{sec}$, which corresponds to an energy spread of

$$\Delta E = 2 E_o \frac{\Delta t}{t_o} = 2.6 \text{ meV} \quad . \quad (4)$$

We estimate a counting rate of 20 counts/sec for a cross section of 0.01 barns and a sample of 10^{23} atoms.

The same experiment could be performed by using the general purpose time-of-flight spectrometer (reference design by Sinha) with a polarized proton filter in the incident beam. In this case we estimate a counting rate of 1600 counts per sec assuming a filter transmission of 0.25. The filter technique thus might give a higher intensity, but would have a lower polarizing efficiency and a much higher cost.

C. MEDIUM ENERGY MAGNONS: THE CONSTANT-Q SPECTROMETER

Magnon and phonon experiments with medium range energy transfers ($\sim 50 \text{ meV}$) and scattering vectors ($\sim 4 \text{ } \text{\AA}^{-1}$) form a large part of the field at present covered on reactors with triple-axis spectrometers. Here we consider a spectrometer allowing this type of experiment, including the opportunity of performing constant \vec{Q} scans, on a pulsed source.

The spectrometer principle is outlined in Fig. 3. The Marx analyzer defines the scattered wavevector k_1 , and hence the scattered neutron flight time t_1 . Time-of-flight analysis of all counters, allows the incident flight time t_o , and hence the incident wavevector k_o , to be determined. Setting up axes parallel and perpendicular to k_o , the components of the scattering vector \vec{Q} are

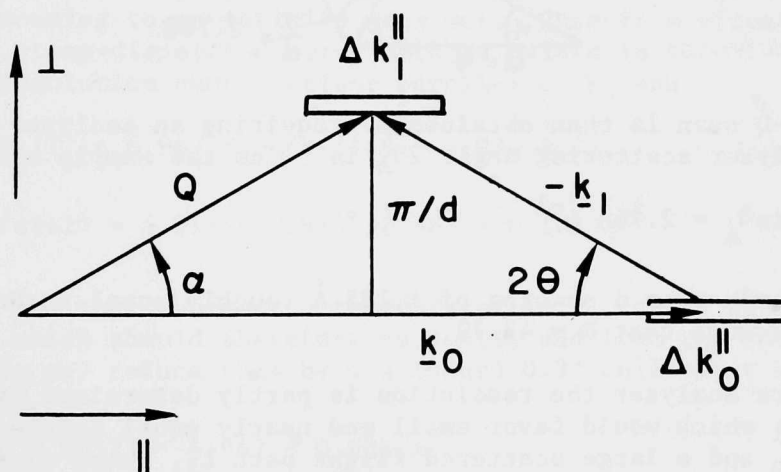
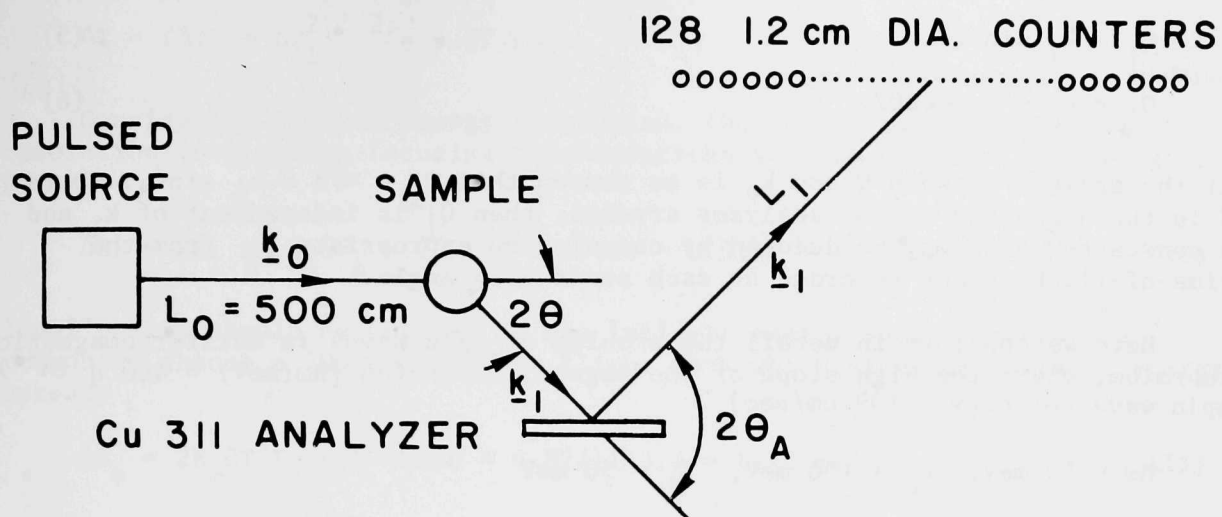


FIGURE 3

A TIME OF FLIGHT MARX SPECTROMETER

$$Q_{\perp} = k_1 \sin 2\theta \quad (5)$$

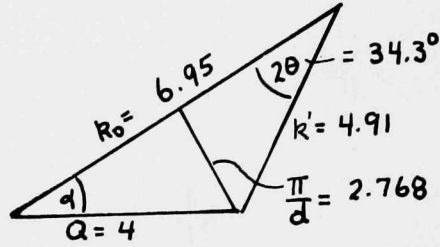
$$Q_{\parallel} = k_0 - k_1 \cos 2\theta \quad (6)$$

If the angle α between \vec{Q} and \vec{k}_0 is so chosen that $Q_{\perp} = \pi/d = k_1 \sin \theta_A$, where d is the d spacing of the analyzer crystal, then Q_{\perp} is independent of k_1 and a constant- Q scan may be deduced by choosing an appropriate Q_{\parallel} from the time-of-flight scans recorded at each scattering angle.

Here we consider in detail the problem of spin waves in antiferromagnetic chormium, where the high slope of the magnon dispersion [$\hbar\omega(\text{meV}) = 400 q (\text{\AA}^{-1})$], spin wave velocity $\sim 10^6 \text{ cm/sec}$

$$\hbar\omega = 50 \text{ meV}, \quad E_0 = 100 \text{ meV}, \quad E_1 = 50 \text{ meV}$$

$$Q = 4 \text{ \AA}^{-1}, \quad k_0 = 6.947 \text{ \AA}^{-1}, \quad k_1 = 4.913 \text{ \AA}^{-1}$$



The constant- Q scan is then obtained by requiring an analyzer crystal such that the analyzer scattering angle $2\theta_A$ is twice the sample scattering angle

$$\frac{\pi}{d} = k' \sin \theta_A = 2.768 \text{ \AA}^{-1} \quad (7)$$

This corresponds to a d spacing of 1.135 \AA roughly equal to Cu 311, and a crystal angle such that $\alpha = 43.79^\circ$.

In a Marx analyzer the resolution is partly determined by the angular divergence α_1 which would favor small and nearly equal sample and counter dimensions d_0 and a large scattered flight path L_1 , where $\alpha_1 = \sqrt{2} d_0/L_1$. In a time-of-flight spectrometer this must be balanced against the time spread of scattered neutrons which is minimized by reducing L_1 . We assume the compromise $d_0 = 1.2 \text{ cm}$ sample and counter diameters, and $L_1 = 139.1$ so that $\alpha_1 = \sqrt{2} \times 1.2/139.1 = 0.0122 (0.7^\circ)$. Assuming a mosaic spread in the analyzer small compared with this, the analyzer angular acceptance $\Delta\theta_A$ is equal to α_1 , and the time spread of scattered neutrons is

$$\Delta T_1 = T_1 \cot \theta_A \Delta \theta_A = 450.8 \times 1.466 \times 0.0122 = 8.06 \text{ } \mu\text{s} \quad (8)$$

This is only slightly larger than the moderation time of 0.1 eV neutrons which is given by $\Delta T_0 = 1.8/\sqrt{0.1} = 5.69 \text{ } \mu\text{s}$. The overall time pulse broadening which will ultimately determine the incident energy resolution is therefore

$$\Delta T = (\Delta T_0^2 + \Delta T_1^2)^{1/2} = 9.87 \text{ } \mu\text{s} \quad . \quad (9)$$

Considering now the energy resolution, the scattered neutron energy resolution (neglecting focusing considerations for the present) will be given by

$$\Delta E_1 = 2E_1 \cot\theta_A \Delta\theta_A = 2 \times 50 \times 1.466 \times 0.0122 = 1.79 \text{ meV} \quad (10)$$

Ideally this should be matched to the incident energy spread. For this reason we choose a 500 cm incident flight path which gives an incident energy spread

$$\Delta E_0 = 2E_0 \Delta T/T_0 = 2 \times 100 \times 9.87/1145.7 = 1.72 \text{ meV} \quad (11)$$

The final overall energy resolution is therefore

$$\Delta E = (\Delta E_0^2 + \Delta E_1^2)^{1/2} = 2.45 \text{ meV} \quad . \quad (12)$$

Next considering the q resolution, we assume again a negligible analyzer mosaic, and also that the incident beam collimation (α_0) is not a limiting factor. Both $\vec{\Delta k}_1$ and $\vec{\Delta k}_0$ are then parallel to \vec{k}_0 as shown. The resolution function is therefore of negligible extent perpendicular to \vec{k}_0 , and we have a situation of focusing to an infinite gradient. This is a situation most appropriate to a steep dispersion curve such as exists in chromium. The extent of those resolution contributions parallel to k_0 are

$$\Delta k_0^{\parallel} = k_0 \Delta T/T_0 = 6.947 \times 5.69/1146 = 0.034 \text{ } \text{\AA}^{-1} \quad (13)$$

$$\Delta k_0^{\parallel} = k_1 \alpha_1 / \sin 2\theta = 4.913 \times 0.0122/0.563 = 0.106 \text{ } \text{\AA}^{-1} \quad (14)$$

Next considering the contributions perpendicular to \vec{k}_0 , these arise from the analyzer mosaic, which should therefore be small, and from the incident beam divergence α_0 . We may reduce this by a standard 0.3° collimator so that

$$\Delta k_0^{\perp} = \alpha_0 k_0 = 0.00523 \times 6.947 = 0.036 \text{ } \text{\AA}^{-1} \quad . \quad (15)$$

Thus the resolution ellipsoid is compressed parallel to \vec{k}_0 and has dimensions $0.111 \times 0.036 \text{ } \text{\AA}^{-2}$ in q and 2.45 meV in energy. The time average neutron flux observed at a 5 m flight path with 0.3° incident collimation is

$$\phi = \frac{0.64 \times 10^{13} \times (0.005)^2}{0.1} \times \frac{0.00523}{0.02} = 4.2 \times 10^8 \text{ ns}^{-1} \text{ eV}^{-1} \text{ cm}^{-2} \quad (16)$$

With the standard cross-section and sample [$b = 1$ in units of 10^{-12} cm, $N = 10^{23}$ atoms] $Nb^2 = 10^{-1}$ the flux at the counter is determined by its solid angle at the sample, $\Delta\Omega$ and the accepted incident energy window ΔE_0 .

If we assume a $5 \times 1.2 \text{ cm}^2$ counter area, and a crystal reflectivity $\eta = 30\%$, the count rate is of the order

$$\begin{aligned}
 N &= \phi \cdot Nb^2 \cdot \Delta\Omega \cdot \Delta E_o \cdot \eta \\
 &= 4.2 \times 10^8 \times 0.1 \times \frac{5 \times 1.2}{(139.1)^2} \times 0.00172 \times 0.3 \\
 &= 6.7 \text{ cts s}^{-1}
 \end{aligned} \tag{17}$$

This is an ample count rate for the experiment, since for chromium the magnetic scattering length p is $\sim 0.17 \times 10^{-12} \text{ cm}$, so that $b^2 \rightarrow p^2 \sim 0.03$. We thus achieve for this point in the scan 12 cts min^{-1} .

To consider the useful length of the scan we consider an array of 128 counters arranged as shown in a time parallel to the incident beam with spacing 1.2 cm . If these are arranged to cover sample scattering angles between 21.4° and 63° , the scan covers energy transfers between 18.3 and 85.9 meV . Near our central energy transfer of $\hbar\omega = 50 \text{ meV}$, adjacent counters occur at energy transfer intervals of order 0.5 meV .

D. HIGH ENERGY MAGNETIC INELASTIC SCATTERING

The observation of interband transitions in the transition metals, both paramagnetic and ferromagnetic, by means of neutron inelastic scattering would initiate a new research area which potentially yields more information on the electronic structure of such materials than does the study of electromagnetic interband transitions. A necessary requirement for such observations is an intense source of neutrons of energy greater than 1500 meV . Typically energy transfers ΔE for such transitions lie in the range $200 \text{ meV} < \Delta E \lesssim 300 \text{ meV}$ and the wavevector should sweep the entire Brillouin zone in a range near $Q \sim 4 \text{ \AA}^{-1}$.

The observation of interband transitions is summarized by the material property, $\chi_2^{\alpha\beta}(\vec{q}, \omega)$, its generalized susceptibility. This susceptibility depends upon matrix elements of the orbital and spin scattering operators and the energy density of interband transitions for given vector \vec{q} . With certain exceptions, this generalized susceptibility is thought to be a smooth function of the order of several electron volts wide. Structure exists on top of the smooth function, and such structure is envisioned to be comparable in width to that observed in optical reflectance measurements, or 200 meV wide. To observe such structure, a resolution of 10 meV is required. Energy transfer of 400 meV requires an incident energy $\gtrsim 1500 \text{ meV}$ for $Q \sim 4 \text{ \AA}^{-1}$.

Examples of $q = 0$ and $q \neq 0$ transitions are shown in Fig. 4. The $q = 0$ transitions give rise to the orbital contribution to the paramagnetic susceptibility, and such contributions are estimated to be appreciable in the metals such as V, Nb, Cr, and Zr ($10^4 \chi_{\text{orb}} = 1.78, 0.98, 1.35, \text{ and } 1.0 \text{ emu/mole}$, respectively). This orbital susceptibility is proportional to

$$| \langle F | L_{\text{op}} | I \rangle |^2 / \Delta E \text{ where } F \text{ and } I \text{ are final unoccupied and initial occupied states, } L_{\text{op}} \text{ is}$$

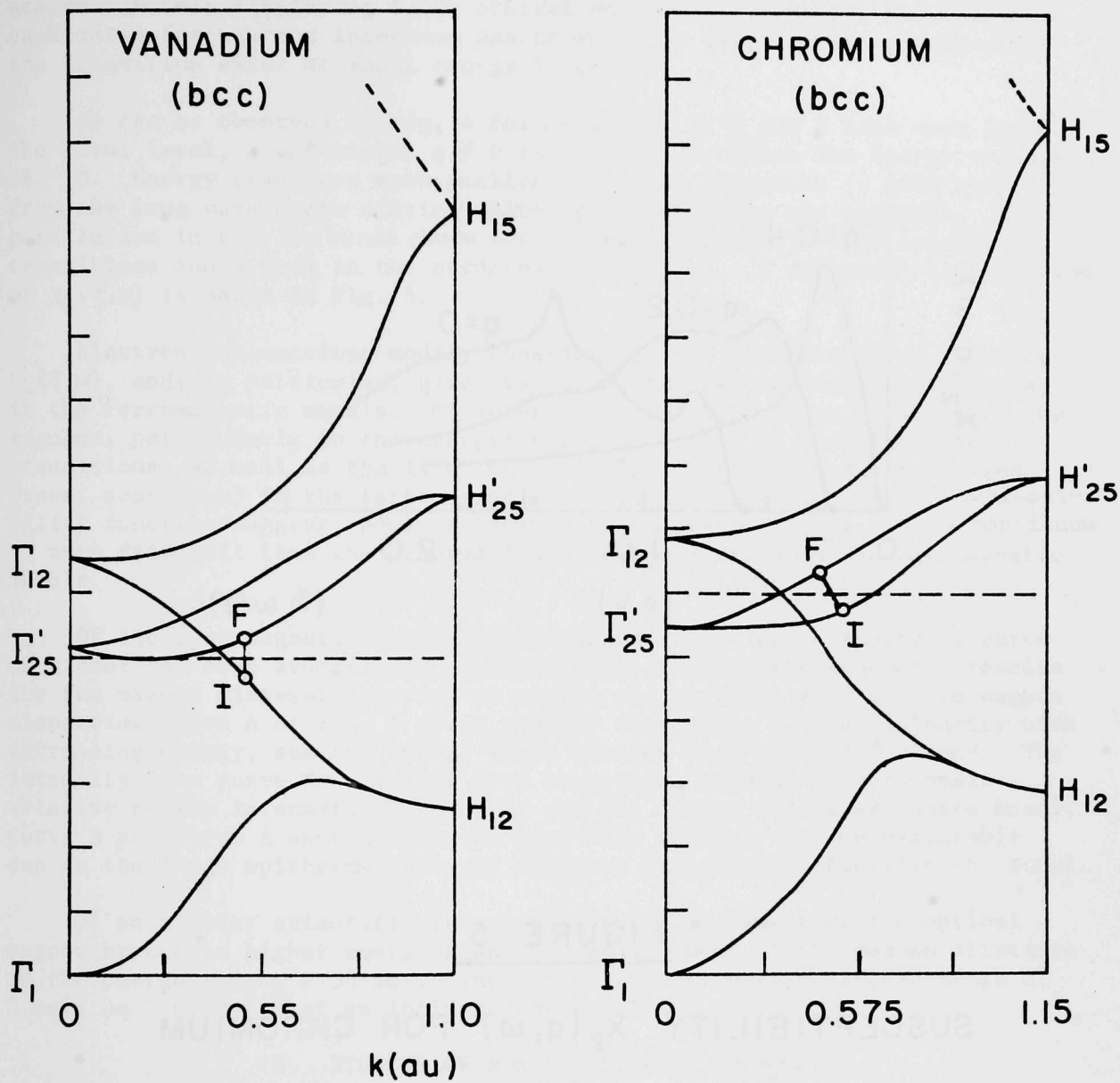


FIGURE 4

POSSIBLE INTERBAND TRANSITIONS

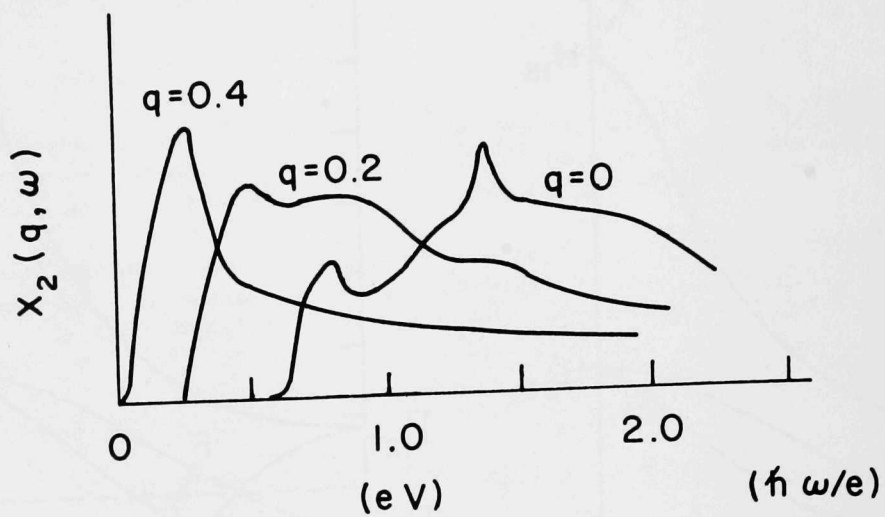


FIGURE 5

SUSCEPTIBILITY $X_2(q, \omega)$ FOR CHROMIUM

the orbital angular momentum operator, and ΔE is the energy different between final and initial states. The average is taken over all initial and final states. Metals displaying large orbital susceptibilities are thus prime candidates for neutron interband spectroscopy as large matrix elements for the transition exist at small energy transfer.

As can be observed in Fig. 4 for chromium, as I and F both move toward the Fermi level, a wavevector $q \neq 0$ is reached for which the energy transfer $\Delta E = 0$. Energy transfers much smaller than that estimated (≈ 1400 meV) from the long wavelength static orbital susceptibility are possible. The parallelism in the two bands shown for chromium suggests a high density of transitions and a peak in the structure of $\chi_2(\vec{q}, \omega)$. A schematic illustration of $\chi_2(\vec{q}, \omega)$ is shown in Fig. 5.

Electron interactions modify considerably the susceptibility function $\chi_2(\vec{q}, \omega)$, and, in particular, give rise to collective excitations, magnons, in the ferromagnetic metals. Of interest is the dispersion relation of the magnons, particularly in the energy region near that of the "interband" transitions, as well as the lifetime of interband transitions (so-called Stoner continuum) in the ferromagnetic metals. The sum rule on the susceptibility function suggest, however, that the observation of the Stoner continuum is more difficult than the observation of the continuum in the paramagnetic metals.

Of the ferromagnetic transition elements, the magnon dispersion curve of nickel has been studied most extensively. Preliminary schematic results for the magnon dispersion curves of nickel are shown in Fig. 6. The magnon dispersion curve A of Fig. 5 gives rapidly decreasing neutron intensity with increasing energy, and the energy width becomes very broad at 140 meV. The intensity from curve B, possibly from an optical magnon, is very small relative to the intensity of curve A, and the energy widths are quite broad. Curve B and curve A above ≈ 100 meV have only recently become measurable due to the large epithermal flux of neutrons from the ILL facility (H. Mook).

Of particular scientific interest is the measurement of the optical magnon branch to higher energies and the measurement of the magnon lifetimes in the energy regime > 50 meV. The latter sets resolution requirements of 5 meV, $\Delta q \sim 0.03 \text{ \AA}^{-1}$ at an incident energy of 400 meV.

E. STUDIES OF NON-EQUILIBRIUM PHENOMENA

1. First Order Phase Transitions

The time scale of the relaxation of a system subjected to a sudden perturbation (temperature, pressure, magnetic field) may vary in a range comparable with the pulse rate of IPNS. For instance Dillon, et al.,¹ have made motion pictures of the nucleation of domains in FeCl_2 and DAG by magneto-optical methods. An example of the measurement of relaxation on the time scale of minutes to hours with neutrons has been the response of the magnetic Bragg peak in Ni_3Mn near the order-disorder transition following the application of a small temperature change.²

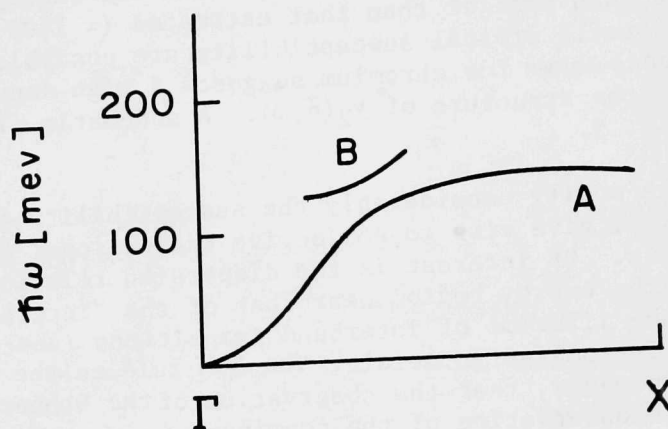
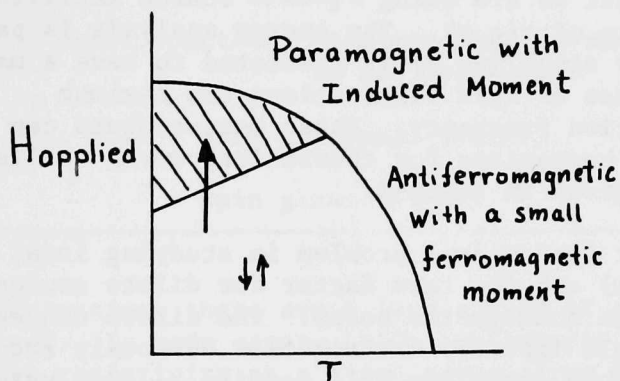


FIGURE 6
PRELIMINARY MAGNON DISPERSION
CURVE FOR NICKEL

Two regimes are envisaged (a) when the relaxation time is much less than the time between pulses and (b) when the relaxation time is much greater than the time between pulses. In case (a) the field, say, is pulsed at the same rate as the neutron source but the time lag between perturbation and neutron burst is varied. In case (b) the field is pulsed once and the time evolution of the signals in the next, say, 10 neutron bursts is followed. This pattern is then repeated.

As a specific example we might wish to study the nucleation of paramagnetic domains in antiferromagnetic FeCl_2 in a magnetic field. Sudden application of a moderate magnetic field (8 - 10 kG) takes the system into the dashed two phase region, nucleating "paramagnetic" domains in an antiferromagnetic "sea". Two kinds of measurements are envisaged as a function of time: small angle scattering to obtain size and shape information about the domains and Bragg intensity and peak profile measurement to obtain the volume of the domains and size information. The effects would obviously depend on the size of the perturbing applied field and also its time deviation. Neutrons would be quite necessary for such studies in bulk material.



Other examples where the perturbations would be easily matched to the pulsing of the instrument include the time dependent response to very high magnetic fields (although there is a problem getting the beam in and out of the magnet), shock waves and laser light. The dynamics of martensitic transformations might be followed in a favorable case.

2. High Pulsed Magnetic Fields

A pulsed neutron source permits one to extend the range of applied magnetic fields in neutron scattering experiments by a factor of at least 2.5, and perhaps even by a factor of 5 over those currently available at steady state sources (~ 100 kG). These high fields (250 - 500 kG) can be achieved by pulsed operation of solid, machined, helical Be-Cu coils as described by Foner.³ For the highest fields it appears necessary to cool the coils to 77 K. By synchronous pulsed operation of the field coils, phase-locked to the proton accelerator envisaged for IPNS, the field need only be on when the pulsed neutrons are striking the sample, thus minimizing the average heat load. Depending on the cooling capacity available, the time that the magnetic field is on can be lengthened to make use of a wider energy spread of the incident neutron beam. This, of course, depends on the distance between the moderator

and the sample. There are numerous coil geometries which should be considered depending upon the application. A coil is described by Foner with a 3/4" bore ($\sim 3"$ long) which produces 550 kG with a pulse half period of 500 μ s for a discharge of 75 kJ. Of course, the energy required is lowered for shorter pulse widths. Difficulties in getting the beam in and out of the coil will vary depending upon the application. For low angle scattering experiment in which a longitudinal field (along beam line) is acceptable, there is no difficulty. To obtain fields transverse to the incident beam it may be necessary to send the beam through the coil material. Split coil (Helmholz pair) operation should be considered.

Important applications of such high fields in neutron scattering investigations of magnetism are numerous, especially in weakly susceptible materials, and in conjunction with the study of magnetic phase diagrams.

F. CONFIGURATIONAL FLUCTUATIONS (BETWEEN STATES OF DIFFERENT SPIN)

IPNS is uniquely suited for investigating moment formation in 3d and 4f metals and compounds and the configuration fluctuations found in some 4f systems. The fact that we are using a pulsed source facilitates making time-of-flight measurements of $\chi(q, \omega)$. The energy analysis is particularly important for these systems since $\chi(q, \omega)$ is predicted to have a maximum at $\omega \neq 0$. For example in the case of spin fluctuations the maximum predicted to occur at the fluctuation frequency. These measurements can serve as both a guide and a standard comparison for theoretical work. A list of problems of current interest is given in Table 2.

Perhaps the most fundamental problem in studying local moment formation is to determine $\chi(q, \omega)$ and the form factor for dilute concentrations of magnetic impurities in nonmagnetic hosts. The dilute concentrations required to determine the single impurity contribution obviously require high neutron fluxes. Further in some cases one will require a large value of magnetic field ($H \approx 100-400$ kG) to saturate the spins in order to separate the magnetic from the nonmagnetic part of the scattering.

In the case of spin fluctuations $\chi''(\omega) = \mu^2 \omega / \pi (\omega^2 + \omega_{5f}^2)$. This leads to

$$\frac{d\sigma}{d\Omega d\omega} (\omega_{5f}) \approx \frac{6 \times 10^{-5}}{\omega_{5f}} \left[\frac{\text{barns}}{\text{steradian eV}} \right]$$

for a sample with 10^3 ppm impurities ($\mu = 3 \mu_B$). For systems like Al:Mn or Y:Ce where the incoherent scattering is small it should be possible to perform these experiments.

Energy analysis will also pay a large role in studying configurational fluctuations. In this case perhaps the best systems to study are Ce and SmS under pressure and alloys such as CeTh for which the transition occurs as a function of T. In these cases the high fluxes and pulsed nature of the source allow one to use pulsed magnetic fields to remove the incoherent scattering and hence obtain high resolution spectra.

Table 2. Configurational fluctuation problems of current interest.

- -4f materials (Ce + cpds)
 - Configurational transitions (spectroscopy) in metals (Eu, Sm, La)
 - Spin fluctuations 5f metals and compounds
 - paramagnets
 - { dilute 3d + 4f in nonmagnetic host $\chi(q, \omega)$
 - { requires high intensity source, ω structure at various q
 - { enhanced 3d in enhanced host (4d, 5d) magnetic cluster, giant moments
 - Concentrated 3d in nonmagnetic-spin glasses (Cu-Mn, Au-Fe)
 - { $\chi_{zz}(q)$ polarized neutrons, polarization analysis
 - { $\chi_{\pm}(q, \omega)$ - exact spectrum, requires high intensity source to get resolution
 - Cone 3d in mag. host - weak ferromag. (Pd-Fe)
 - spin glass (Pd-Mn)
-

At higher concentrations there are a large class of systems which are very poorly understood. In some cases, La:Ce for example, interaction effects appear in the resistivity at a given temperature at an order of magnitude lower concentration than they do in the susceptibility. Clearly measuring $\chi(\vec{q}, \omega)$ would help to resolve this problem. Similar measurements with polarization analysis on such spin glass systems as Cu:Mn and Au:Fe would be very helpful in resolving the type of ordering that occurs in these systems. Other types of systems that would be interesting to investigate and typical examples are: giant moment systems and weak ferromagnets (Pd:Fe).

G. TOPICS DISCUSSED BRIEFLY

1. Experiments requiring high energy incident neutrons ($E_0 > 150$ meV)
 - Optic magnons; giant moment problem, e.g., Pd(Fe)
 - Amorphous magnetic materials
 - Crystal field levels, dynamic charge transfer, valence fluctuations
 - Highly absorbing magnetic materials
 - Elastic diffuse scattering (quasielastic approximation need low λ_0)

- Form factor anisotropy at large wavevectors (Q up to say 60 \AA^{-1})
2. Experiments requiring the pulsed nature of the source
- Phase diagram, higher order critical phenomena (high B, high P)
 - Weakly susceptible materials, diamagnetism (high B)
3. Experiments requiring highest flux attainable
- Diffraction on very small samples (5 mg or so), magnetic precipitates, rare materials, radioactively transmuting samples, etc.
 - Spin glass alloys, e.g., Cu(Mn), Au(Fe), etc.
 - High resolution experiments on magnons, lifetimes, 2-magnon excitations
 - Surface magnetism
 - Magnetic experiments in biology (on hemoglobin for example)

H. IPNS EPITHERMAL FLUX-ENERGY-RESOLUTION RELATIONSHIPS

Assume:

Time average total flux at the moderator

$$F = \frac{4 \times 10^{13}}{E} [\text{ns}^{-1} \text{ eV}^{-1} \text{ sterad}^{-1}]$$

$$\text{Moderator pulse widths } \Delta T = \frac{1.8}{E^{1/2}} [\mu\text{s}], E \text{ in eV.}$$

$$\text{Flight path } X = \sqrt{2/m} T E^{1/2} = 1.38 T E^{1/2} [\text{cm}], E \text{ in eV, } T \text{ in } \mu\text{s}.$$

$$\text{Incident pulse resolution } R = \frac{\Delta T}{T} = \frac{1}{2} \frac{\Delta E}{E}$$

Then:

$$\text{Flight path } X = \sqrt{2/m} \frac{\Delta T E^{-1/2}}{R} = \frac{2.5}{R} \text{ cm}$$

Time average flux on specimen

$$\phi = \frac{F}{X^2} = \frac{F m R^2}{2 (\Delta T)^2 E} = 0.64 \times 10^{13} \frac{R^2}{E} [\text{n cm}^{-2} \text{ eV}^{-1} \text{ s}^{-1}]$$

Flux per incident pulse resolution element

$$\phi \Delta E = \frac{F_m R^3}{(\Delta T)^2} = 2 \times 0.64 \times 10^{13} R^3 \text{ n cm}^{-2} \text{ s}^{-1}$$

Example:

Choose $R = 0.0025$ then $X = \frac{2.5}{0.0025} = 1000 \text{ cm}$, $E = 0.1 \text{ eV}$

$$\text{Time average flux } \phi = \frac{0.64 \times 10^{13} \times (0.0025)^2}{0.1} = 4 \times 10^8 [\text{n cm}^{-2} \text{ s}^{-1}]$$

$$\begin{aligned} \text{Flux per resolution element } \phi \Delta E &= 2 \times 0.64 \times 10^{13} \times (0.0025)^3 \\ &= 2.4 \times 10^5 [\text{n cm}^{-2} \text{ s}^{-1}] \end{aligned}$$

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VIII. REPORT OF THE PANEL ON NEUTRON SOURCES AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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A. GENERAL COMMENTS ON INTERCOMPARISONS OF PULSED SOURCES

The total fast neutron production rate from a pulsed neutron source is only a first yardstick for comparing different sources. For use of fast neutrons from a bare target, the proper basis for comparison is the angular current density ($n/\text{cm}^2\text{-ster-eV-sec}$) at the target surface. Similarly the neutronic coupling between neutron producing targets and moderators, depends significantly on the size and material of the source, and the angular current density of slow neutrons from the moderator is not accurately proportional to the total neutron production rate.

Below we discuss some of the considerations to be accounted for in comparing pulsed sources, and in design of sources for various purposes.

B. CONSIDERATIONS IN SELECTING A NEUTRON PRODUCING TARGET

The considerations in selecting the size of and material for a neutron producing target fall into at least three general (and somewhat interrelated) categories:

1. Target neutronics and energy distributions.
2. Direct experimental backgrounds.
3. Target energy deposition.

It is necessary to consider the above three categories essentially concurrently because the engineering can override the neutronic considerations in target design.

C. TARGET YIELDS, TARGET/MODERATOR, AND MODERATOR/MODERATOR NEUTRONIC COUPLING

The neutron yields from bare solid cylinders of ^{238}U targets bombarded by 800 MeV protons in a 15 mm diameter beam spot centered about the target-axis is shown in Fig. 1.¹ Both the total neutron leakage as well as the leakage from the cylindrical surface are depicted as a function of target diameter and height. Note at large diameters that end leakage from the cylinders becomes extremely important. For unreflected target/moderator configurations, the moderators are generally neutronicly coupled to leakage neutrons from the cylindrical surface. The neutron leakage intensity (n/cm^2) from the cylindrical surface decreases with increasing target diameter; this affects the target-moderator neutronic coupling.

The energy spectrum of leakage neutrons from the cylindrical surface of the ^{238}U targets becomes softer with increasing target diameter as shown in Fig. 2.¹ The softening of the spectrum aids in the neutronic coupling between a target and a moderator.

The effect of coupling one 50-mm thick by 150-mm long by 150-mm² high H_2O moderator to a cylindrical target in a single-wing configuration² is illustrated in Fig. 3.¹ The moderator was 10-mm from the circumference of the bare target. The neutron leakage indicated in Fig. 3 is the angle-

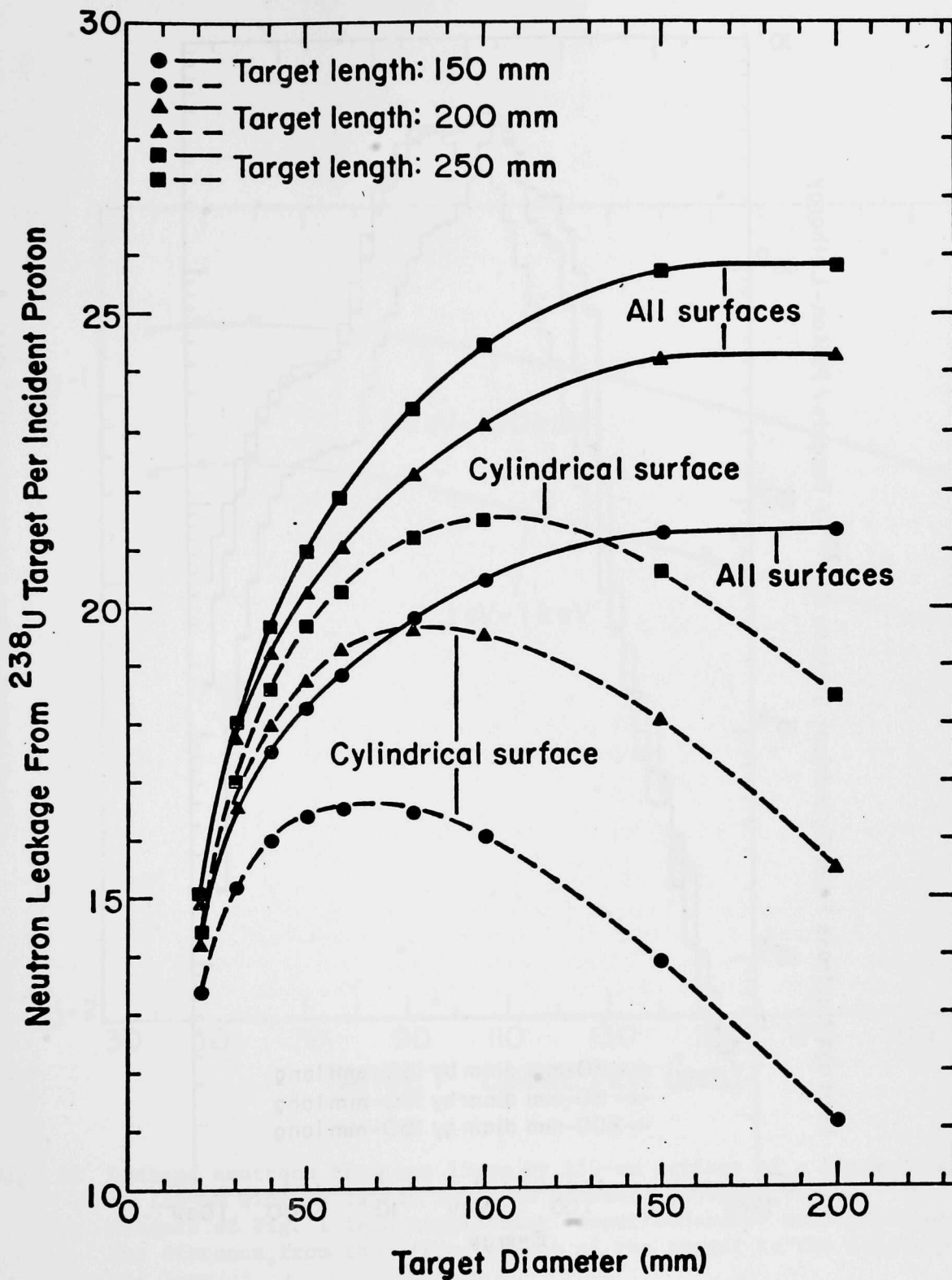


Fig. 1: Total neutron leakage yields and yields from the cylindrical surface for solid cylinders of ^{238}U bombarded with 800 MeV protons. An atom density of 4.73×10^{22} atoms/cm³ was used for the ^{238}U ; a uniform proton beam spot of 15-mm diam centered about the axis of the cylinder was assumed.

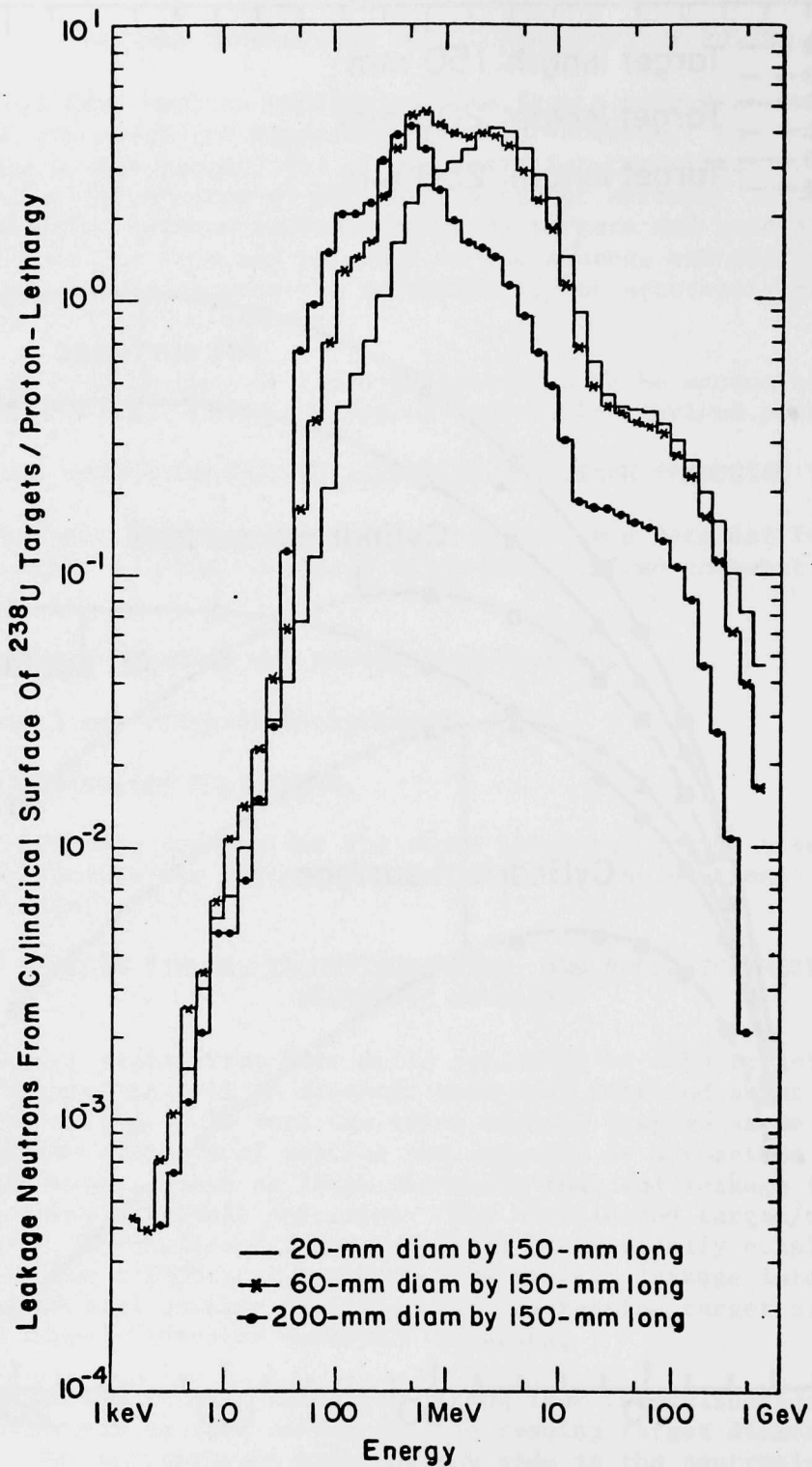


Fig. 2: Neutron leakage spectra from the cylindrical surface of ^{238}U targets. The target conditions were as for Fig. 1.

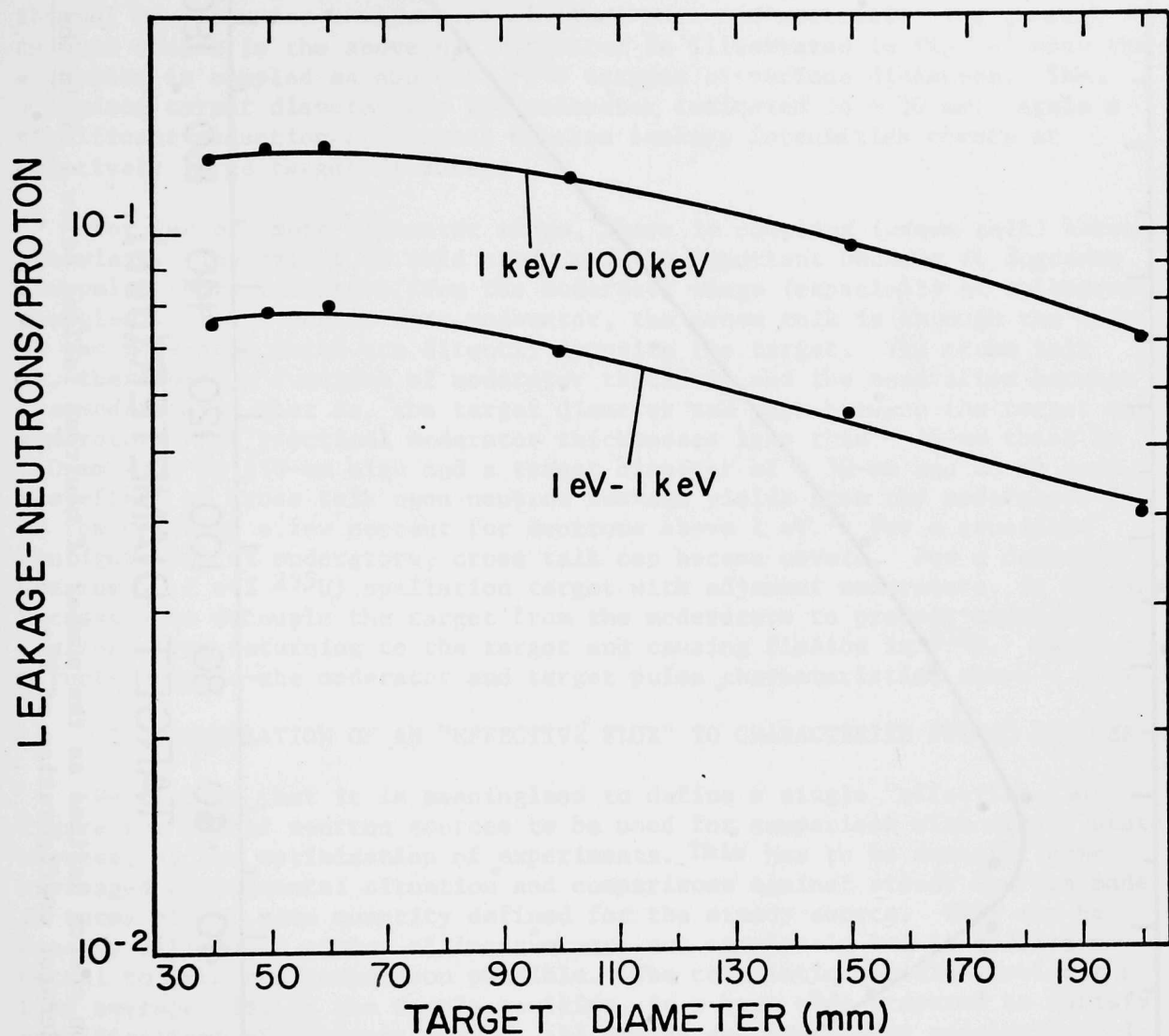


Fig. 3: Leakage neutrons from one 15-mm by 150-mm surface of a 50-mm thick by 150-mm wide by 150-mm long H_2O moderator coupled to the ^{238}U targets of Fig. 1 in a single-wing target/moderator configuration. The distance from the circumference of the target to the moderator was 1-mm.

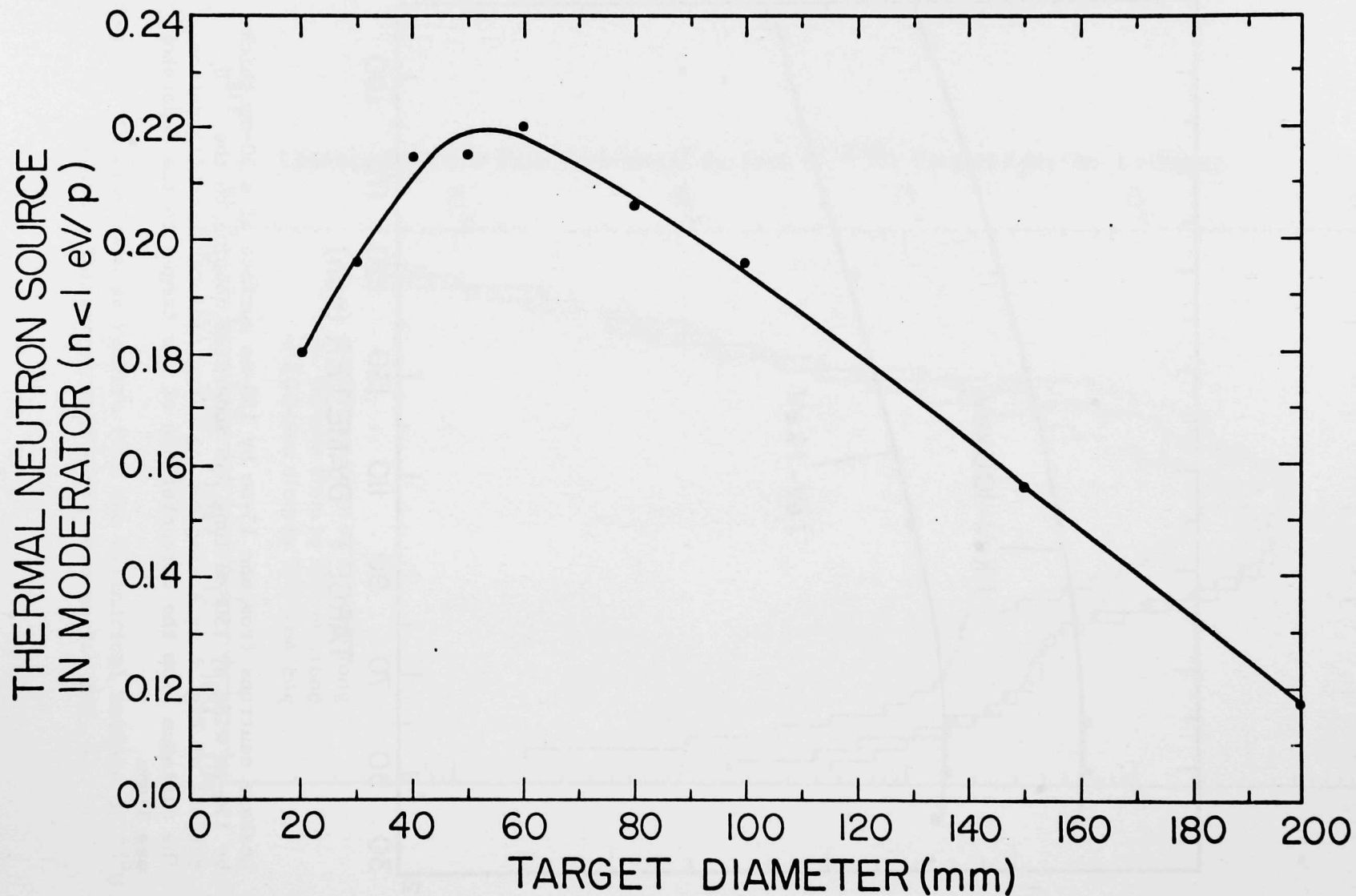


Fig. 4: The thermal neutron source defined as the number of neutrons falling below 1 eV in the moderator for the target/moderator configuration of Fig. 3.

integrated leakage from one 150-mm by 150-mm moderator surface for the energy ranges indicated. Note the significant decrease in leakage intensity as the target diameter increases.

The thermal neutron source in the moderator* should be an indicator of the neutron leakage intensities which could be expected at thermal and sub-thermal energies for a moderator of fixed size and material. The thermal neutron source in the above H₂O moderator is illustrated in Fig. 4¹ when the moderator is coupled as above to ²³⁸U targets of various diameters. The optimized target diameter for the moderator indicated is ~ 50 mm. Again a significant reduction of thermal neutron leakage intensities occurs at relatively large target diameters.

For two or more moderator wings, there is coupling (cross talk) between the wings. The extent of this cross talk is important because it degrades the pulse characteristics from the moderator wings (especially at epithermal energies). For a double-wing moderator, the cross talk is through the ends of the moderator which are directly opposite the target. The cross talk is, therefore, a function of moderator thickness and the separation between the moderators, that is, the target diameter and gaps between the target and moderator. For practical moderator thicknesses less than ~ 50 -mm thick by 150-mm wide by 150-mm high and a target diameter of ~ 30 -mm and 10 mm gaps, the effect of cross talk upon neutron leakage yields from the moderators is on the order of a few percent for neutrons above 1 eV.¹ For a cruciform configuration of moderators, cross talk can become severe. For a depleted uranium (0.2 wt% ²³⁵U) spallation target with adjacent moderators, it appears necessary to decouple the target from the moderators to prevent moderated neutrons from returning to the target and causing fission in ²³⁵U. Such effects degrade the moderator and target pulse characteristics above ~ 1 eV.

D. FORMULATION OF AN "EFFECTIVE FLUX" TO CHARACTERIZE PULSED SOURCES

We suggest that it is meaningless to define a single "effective flux" figure for pulsed neutron sources to be used for comparison with steady state sources, or for optimization of experiments. This has to be done for some envisaged experimental situation and comparisons against steady sources made in terms of the same quantity defined for the steady source. This may be done by defining a number of instruments, not completely but in sufficient detail to make the comparison possible. The calculations could provide the time average flux at the sample position, in a beam once prepared to satisfy specifications of incident energy, and energy and wavevector resolution. In the pulsed case the beam is prepared by pulsing and the source to sample distance is determined by resolution requirements. In the steady state case, the beam in the conceptual instrument might be prepared by crystal monochromation. In the pulsed source case, the analysis might follow that of Windsor and Sinclair.³

With instruments thus conceptually defined, the time average flux at the sample might be computed, and tabulated for various purposes as below (for different classes of measurements the instruments would be different).

* Defined as the number of neutrons falling below 1 eV in the moderator.

Incident Energy	1 eV				0.5 eV			0.1 eV etc.
		Elastic 10^{-3}	K 10^{-2}	Resolution 10^{-1}		10^{-3}	10^{-2}	10^{-1}
Small K $10^{-2} \leq K \leq 1 \text{ \AA}^{-1}$	Inelastic Energy Resolution 10^{-3}				10^{-3}			
	10^{-2}				10^{-2}			
	10^{-1}				10^{-1}			
Medium K $1 \text{ \AA}^{-1} \leq K \leq 10 \text{ \AA}^{-1}$							etc.	
Large K $10 \text{ \AA}^{-1} \leq K \leq 10^2 \text{ \AA}^{-1}$								

Consideration should be given to the variety of choices of moderator design, to the use of neutron guides, as do Windsor and Sinclair, and to the constraints imposed by frame overlap conditions.

We would also suggest that some equivalent experiments should be carried out on both pulsed and steady sources to provide experimental data to give confidence in the approach outlined. A small program of this type exists in the U.K. in relation to the Harwell LINAC. Experiments on 0.3 - 0.5 Å neutrons (K up to 30 Å^{-1} and higher) scattered from amorphous silica and germania have shown that the total scattering spectrometer on the present LINAC is at least as good as the hot source diffractometer (D_4) at ILL. Similar conclusions have been reached for scattering from liquid nitrogen. Given such points of comparison one can scale between different pulsed systems using the time averaged fast flux emerging from the target. This is possible because the moderation problem and methods of utilization are similar in all pulsed systems. It is suggested that a few such experiments be performed.

E. USE OF LOW TEMPERATURE SOURCES OF LONG WAVELENGTH NEUTRONS

In the Workshop panels on Biology and Chemical Structures of Disordered Solids consideration was given to a small K diffractometer and a small K scattering instrument ($\lambda \sim 5$). In both cases instruments were discussed on a time-of-flight basis and in one case it was necessary to envisage a 30 Hz rather than a 60 Hz pulse repetition frequency.

An alternative mode which should be examined is to take 60 s^{-1} of the long pulses ($\Delta\tau \sim 200 - 500 \mu\text{s}$) from a low temperature (methane, 20°K) fully moderated system. Such a sequence of long pulses could be used as a "quasi-steady source" mode with conventional instrumentation and position sensitive detection. Such a mode would make this type of instrument, even though it is not a favorable regime of IPNS, comparable with those generally available. It would be advantageous to use a guide tube. Dr. Watanabe has calculated the flux in an instrument using a statistical chopper on a He-cooled methane source, a 30 M flight path to the chopper and a 85-100 M flight path to the detector, and finds performance at least comparable with the ILL cold source.

F. COMMENTS ON AVAILABLE MODERATOR DATA

A review of moderator studies for pulsed neutron sources was done by D. F. R. Mildner.⁵ In addition to the data recorded in the Draft Proposal for an Intense Pulsed Neutron Source, the following data are available as to cold moderators at $\sim 20 \text{ K}$ and 77 K .

Moderator	Spectrum	Pulse Width	ABS Flux Evaluation
CH ₄	0 (available	0 (partly available)	0 (rough evaluation possible)
C ₂ H ₆	0		0
H ₂	0	0	0
H ₂ O	0		0
C ₆ H ₆ ·3CH ₃	0	0	0
CH ₂ (4 K & 77 K)	0	0	0

The spectra by Inoue, et al.,⁶ measured through reentrant holes in 20 cm dia x 20 cm high cylinders of CH₄, C₂H₆, H₂, and H₂O at various temperatures are shown in Fig. 5. The spectra for a similarly configured moderator of 21.4 K mesitylene (C₉H₁₂) measured by Utsuro⁷ is compared in Fig. 6 with that for 18 K liquid H₂. While these spectra may differ from what would be characteristic of the slab moderators appropriate for a some sources the differences are expected to be small so long as the leakage is similar ($L^2B^2 \sim 0.5$).

Because reference to the work seems not to appear elsewhere, we call attention to the measurements of spectra and pulse widths of liquid para-hydrogen by K chle and W rz,⁸ which are reproduced in Fig. 7.

G. DATA NEEDED FOR FURTHER SOURCE-MODERATOR OPTIMIZATION

The present and immediately prospective data base on source and moderator behavior is considered sufficiently precise for proposals, scoping studies, and for initial design of instruments, but in some cases the data base will be inadequate for instrument refinement and for planning of any optimized facility. The Panel finds it would be highly desirable that some of the questionable points and minor inconsistencies which exist be resolved within the next year or two to allow the new results to be used in facility optimization.

One purpose of such experiments should be to confirm the validity of existing calculational methods which can now yield extensive results for real systems. In particular they can give the energy dependence of the intensity and time jitter of the neutrons emitted from various portions of a moderator surface as a function of energy for neutron energies above ~ 0.5 eV (chemical binding ignored). Particular questions revolve around the accuracy of the treatment in these codes at the highest energies considered (0.5 to 1 GeV). Fission is probably not treated when induced by particles of energy above ~ 20 MeV, and the underlying intranuclear cascade plus evaporation model may not be accurate to 30%.

Another need is for observations of intensity and pulse widths for hopeful moderator materials as a function of output energy and moderator temperature. While the idea is quite satisfactory of patching spectra observed in one

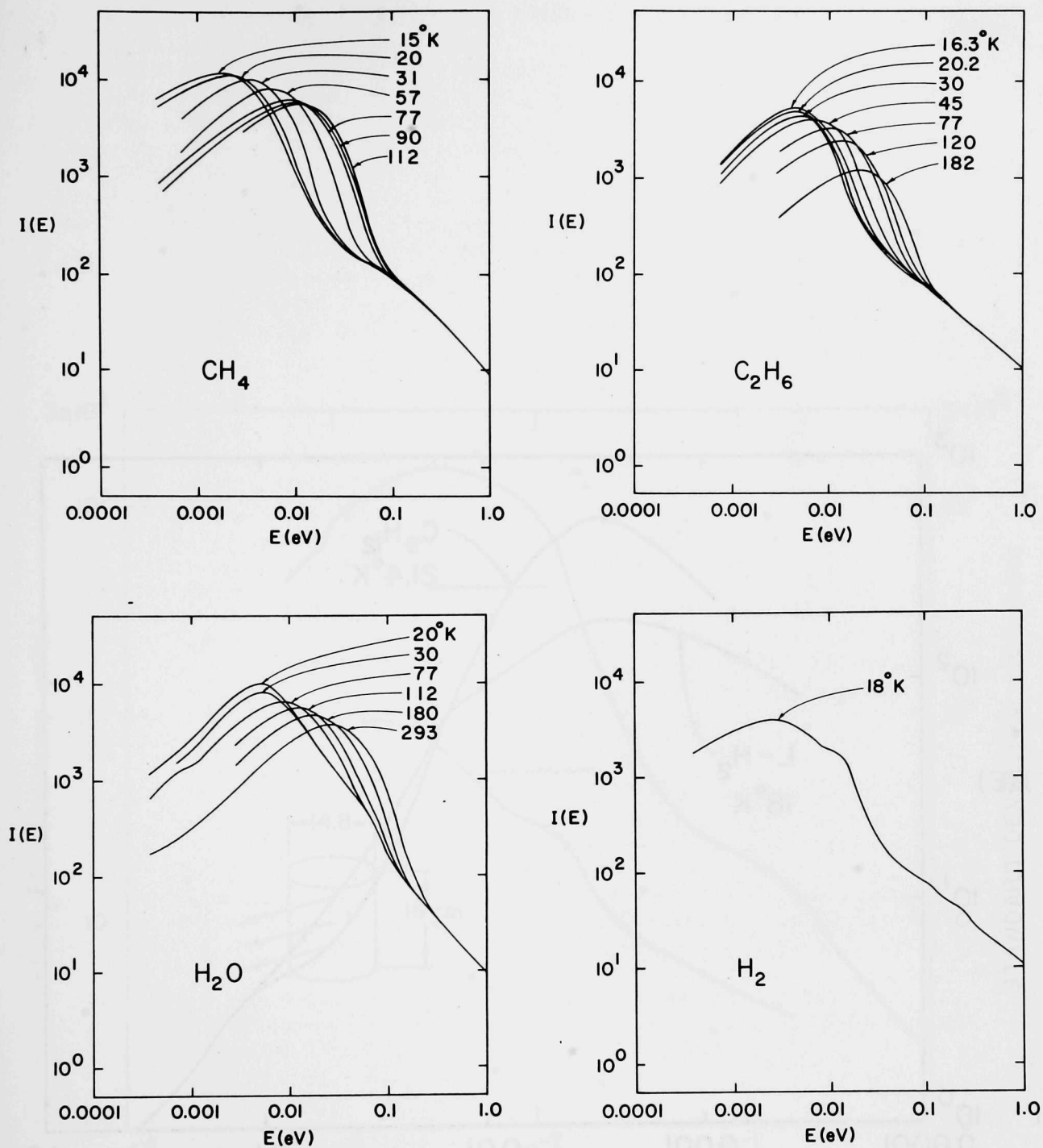


Fig. 5: Measured neutron spectra in liquid and solid methane, liquid and solid ethane, light water, ice and liquid hydrogen.⁶

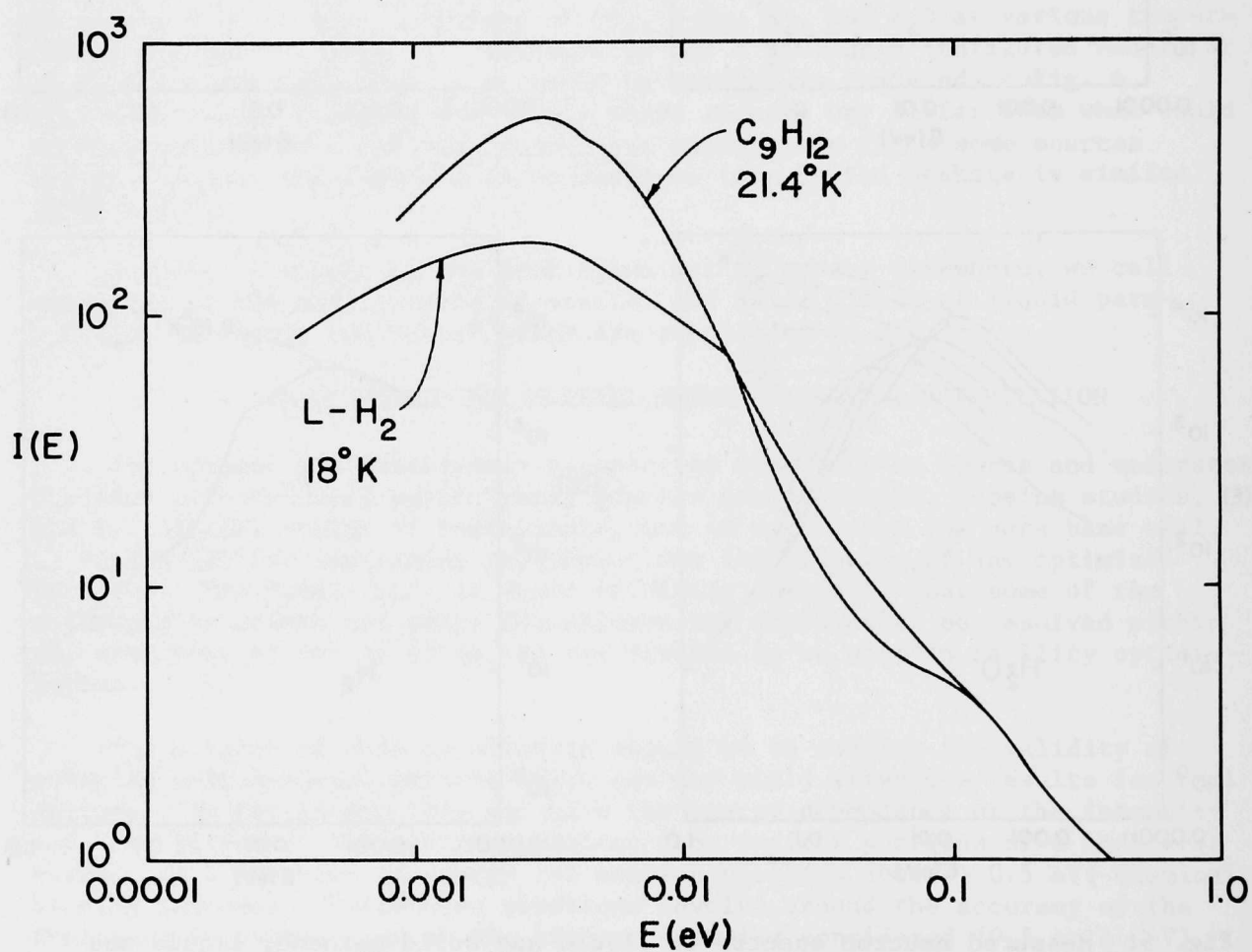


Fig. 6: Neutron spectra from a cold source of mesitylene (1-3-5 trimethyl benzene, C_9H_{12}) at 21.4 K (unpublished measurement by Utsuro) compared with liquid hydrogen at 18 K (Inoue, et al.). The moderators are 20 cm dia \times 20 cm high cylinders, viewed through a reentrant hole.

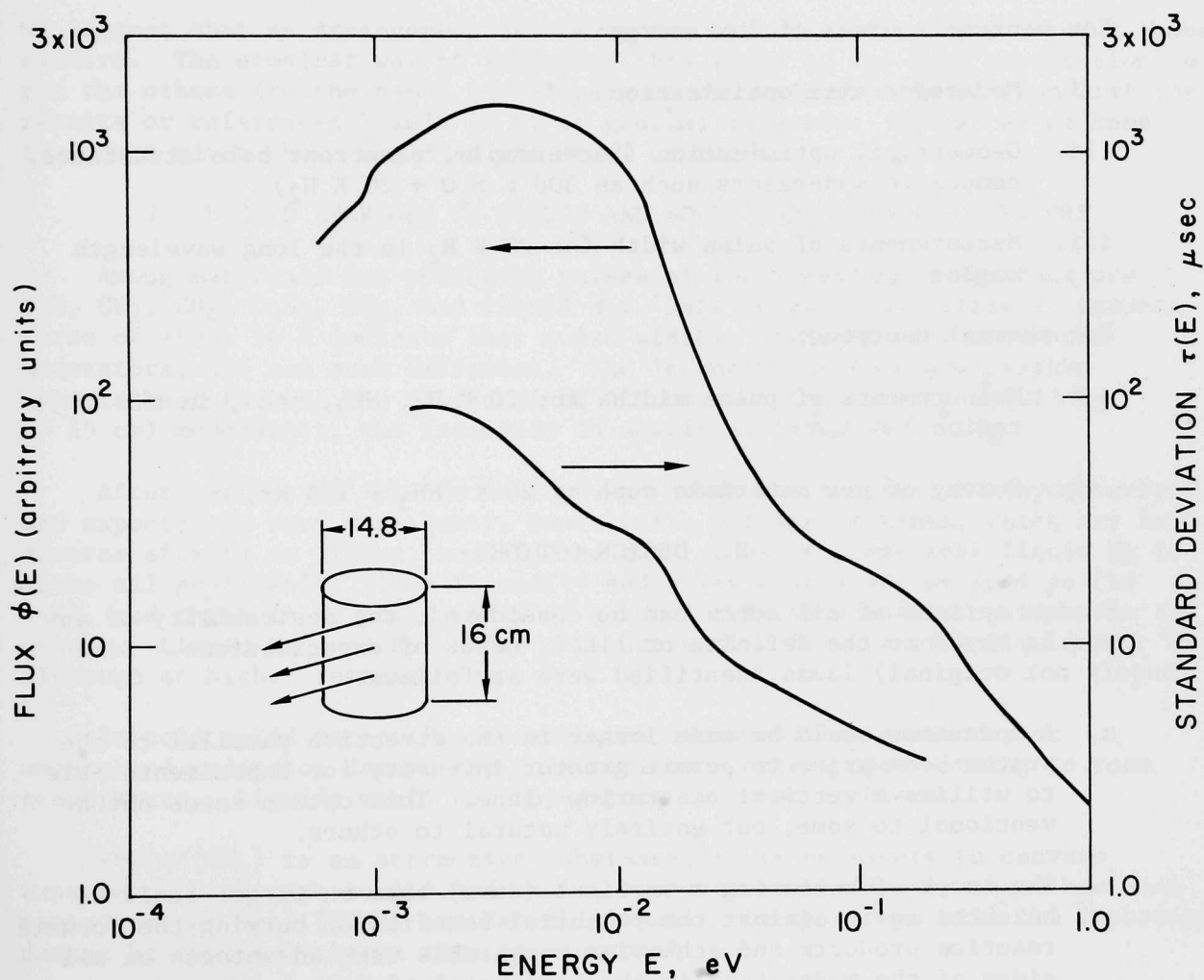


Fig. 7: Neutron spectrum and standard deviation of the pulse from a 97.5% parahydrogen moderator at 20.4 K (unpublished measurements by K  chle). The moderator is a 14.8 cm dia \times 16 cm high cylinder, viewed on the cylindrical surface.

source geometry onto computed intensities for another geometry, it would be reassuring if the spectral observations overlapped the energy region where chemical binding is known to be quite unimportant. (Calculations to lower energies for materials for which the scattering law is known could help bridge this gap.) More urgent are studies showing the pulse widths and intensities to be obtained at 100 meV and below from moderators usable at high-power facilities.

A third domain inviting experimental study is the behavior of the time jitter from reflected moderators at energies below the nominal cutoff energy of the decoupling layer which separates moderator from reflector.

We record here a brief list of data needed for further optimization of slow neutron sources.

For neutron sources of low energy:

- i. Moderator size optimization
- ii. Geometrical optimization (for example, reentrant hole structures, composite moderators such as 300 K H_2O + 20 K H_2)
- iii. Measurements of pulse width for 20 K H_2 in the long wavelength region

For thermal neutrons:

- i. Measurements of pulse widths for 20 K H_2 , NH_3 , etc., in the region $\lambda \sim 1 \text{ \AA}$
- ii. Survey of new materials such as 20 K (NH_3 + 17% Kr)

H. DESIGN OPTIONS

Design options of all sorts can be considered; the desirability of any of these is keyed to the definite or likely needs of experimenters. Some (surely not original) ideas identified were as follows:

- a. A moderator could be made longer in the direction parallel to the proton beam axis, to permit greater intensity for experiments able to utilize a vertical scattering plane. This option seems unconventional to some, but entirely natural to others.
- b. The costs of achieving a vertical (down) beam on target could be balanced again against the potential benefits of burying the forward reaction products and achieving comparable user advantages on all sides of the moderator (in the horizontal plane).
- c. Perhaps all moderators should be chilled, except when some specific experiments requires a "hot" (room temperature) source.
- d. If experimental samples or equipment can afford to "look at" the spallation source, would a moderator configuration more like that now postulated at LASL-WNRF give improved performance?

I. NEUTRON SOURCE AND MODERATOR INFORMATION POOL

From our discussion it has emerged that there are a number of centers actively interested in moderator materials and designs for use with pulsed systems. They include:

Argonne National Laboratory -- Dr. J. M. Carpenter
Los Alamos Scientific Laboratory -- Dr. R. G. Fluharty
Oak Ridge National Laboratory -- Dr. R. W. Peelle
Harwell, U.K. -- Dr. C. G. Windsor
Rutherford Laboratory, U.K. -- Dr. L. C. W. Hobbs
Tohoku University, Japan -- Dr. N. Watanabe
K.E.K. National Laboratory of High Energy Physics -- also Dr. Watanabe
of Tohoku

We suggest that an arrangement be set up for circulating reports between these centers. The simplest way of achieving this would be for each institution to put the others (to the named individual) on an automatic circulation list for reports or references likely to be of general interest. As far as we know the names given above would be the local contacts.

J. LIQUID HYDROGEN VS LIQUID AND SOLID HYDROCARBON MODERATORS

Among materials for producing pulses of long-wavelength neutrons are H_2O , CH_2 , CH_4 , C_2H_6 , NH_3 , and liquid H_2 . Data on these materials at temperatures of about 20 K indicate that pulse widths, at least for unpoisoned moderators, are not much different. The intensity at long wavelengths appears best for CH_4 , in thickness of ~ 5 cm, but for thicker liquid H_2 (~ 15 cm) moderators, the intensity is nearly as large as in CH_4 .

After periods of intense neutron and gamma irradiation, the hydrocarbons are expected to form some heavy, nonvolatile polymer residues, which may have adverse effects on piping, containers, etc. Thus it seems that liquid H_2 is above all preferable, since intensity and pulse widths are as good as the best, and there is no possibility of accumulating nonvolatile residues in the system. Liquid NH_3 and H_2O share this attractive feature with liquid H_2 , although at higher temperatures.

In cold solid materials, accumulated decomposition products, or unannealed radiation damage may pose serious problems upon warming to room temperature. Liquids would not be subject to these problems.

While (CH_2) is an attractive substance, it is necessary to canvass experience on damage to its moderating properties. Evidently there has been experience at Geel (BCMN) and Harwell linacs. In this case will the important damage be caused by gamma rays or neutrons?

K. PRELIMINARY EVALUATION OF COLD NEUTRON SOURCE INSTALLED AT IPNS

If we can install a cold source, for example 20°K CH₄ with an appropriate size at the moderator position "A" or "C" (high intensity position) (see the Draft Proposal for an Intense Pulsed Neutron Source), we can expect the cold neutron flux, the peak value of which is higher than, or at least comparable to, the existing ILL cold flux.

Using the spectrum for 20°K CH₄ measured by Inoue (Fig. 5) and normalizing it at 1 eV to the value $I = 3.5 \times 10^{-4}$ n/ster/eV/n_s (which is strictly not proper because this value applies for a $10 \times 10 \times 5$ cm CH₂ moderator), we can estimate a 4π equivalent peak cold neutron flux at moderator surface, $\hat{\phi}_{4\pi}(E)$ as follows,

$$\hat{\phi}_{4\pi}(E) = \frac{4\pi S_n}{A \tau(E)} 3.5 \times 10^{-4} I_c(E)$$

where

$$S_n = 1.5 \times 10^{15} \text{ n}_f/\text{pulse}$$

$$A = 100 \text{ cm}^2$$

$$\tau(E): 150 \sim 200 \text{ } \mu\text{s for } 4 < \lambda < 8 \text{ } \text{\AA}$$

$$I_c(E): \text{ measured spectrum on 20 K CH}_4 \text{ (Fig. 5)} \\ \text{normalized to 1.0 at 1 eV.}$$

The value of $\hat{\phi}_{4\pi}$ at 6 Å, for example, reaches to 3×10^{17} n/cm²sec/eV with the measured value of $I_c(6 \text{ } \text{\AA}) = 1000$, and with the assumed value of $\tau(6 \text{ } \text{\AA}) = 200 \text{ } \mu\text{s}$.

Further improvements may be possible, but there are some engineering features not accounted for in this idealized calculation which might cancel the improvements. Some advanced techniques should be looked into, such as a correlation chopper which uses cold neutrons of all wavelengths and treats the cold source as a steady source.

L. CALCULATION OF THE INDUCED RADIOACTIVITY OF A STEEL ROTOR IN THE NEIGHBORHOOD OF THE SOURCE

It can be foreseen that for some purposes rotors may be placed close to the neutron source. The activation of these rotors by energetic neutrons is evaluated here as an aid to designs of such installations. The rotor might be located at $\theta = 30^\circ, 90^\circ$ or 150° from the forward proton beam direction.

The numbers of high energy neutrons ($20 < E < 800$ MeV) produced per proton and steradian in the uranium target by 800 MeV protons are as follows:

$\frac{dn}{d\Omega} \frac{n}{\text{sr-proton}}$	θ
0.5	30°
0.03	90°
0.025	150°

We assume the rotor is at 1 meter from the source. Per cm^2 one has thus

$$\Delta\Omega = 10^{-4} \text{ sterad.}$$

With a beam current of

$$I = 3.10^{15} \text{ protons/sec}$$

the flux activating the rotor is thus

$$\phi = I \frac{dn}{d\Omega} \Delta\Omega = 3.10^{15} \times 3.10^{-2} \times 10^{-4} = 9 \times 10^9 \frac{n}{\text{sec cm}^2} .$$

What is the radiation field in rad/hour produced by a body of steel which has been irradiated at saturation? (This is the case after a few weeks or months.) Let us take 1 day cooling time. In Ref. 9 the "danger parameter" of iron irradiated by 600 MeV protons is given (p. 333). It is the radiation field experienced in a cavity embedded in material homogeneously activated by a flux of 10^6 particles/sec cm^2 . We read 46 mrad/h. If one lays one's hand on the activated rotor, one gets radiation from a solid angle 2π instead of 4π in a cavity. Thus the value to be taken is one half of the danger parameter; i.e., 23 mrad/h for 10^6 n/s cm^2 . Now we have 9×10^9 n/sec cm^2 ; i.e., 207 rad/h.

This is if the rotor is in view of the source without material in between. Actually, we might have 0.5 meter of beryllium and 0.5 m of iron, giving 1 high energy neutron attenuation mean free path for the beryllium and 3 for the iron; i.e., an attenuation of $e^4 \approx 55$ and we get for the radiation field of the rotor at contact

$$\frac{207}{55} \approx 3.6 \text{ rad/h} .$$

If the rotor is not big, the radiation level falls rapidly with distance (as the solid angle under which the rotor is seen). We assume that the whole body of a worker gets much less and that the limit is set by the tolerance on the hands, which is 15 rem in one year (or rad in one year in this case). If the radiation dose is received in one that a limit of 9 rem is prescribed.

Consequently a man can be admitted to work on this rotor up to a maximum of 2.5 hours. He will then have received 9/15 of the yearly dose allowable on his hands (60%).

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IX. REPORT OF THE PANEL ON RADIATION EFFECTS AT THE
WORKSHOP ON USES OF ADVANCED PULSED NEUTRON SOURCES

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I. INTRODUCTION

The authors of this report have studied the IPNS radiation effects facility primarily with the aim of exploring the usefulness of this device as a research tool in radiation damage studies. The features of particular interest are the relatively high flux coupled with a low nuclear heating, experimental access, and the distinctive neutron spectrum all of which make this an attractive instrument to pursue fundamental radiation damage studies. The novel feature of a pulsed beam that appears to present unique possibilities for neutron scattering is likely to be less significant to the radiation damage community. By necessity the time average features of the device are the most important and will receive most attention. Moreover, the radiation effects facility is a tool directed towards the fundamental studies, and is not intended nor particularly suitable for selection and qualification of materials for high-flux reactor applications. With these considerations in mind, several research areas in radiation effects were examined.

The general characteristics of IPNS were taken to be as follows:

Pulsed source: 60 Hz repetition rate, 0.5 μ sec pulse width
Flux: $\sim 10^{18}$ n/cm² sec during pulse; $\sim 2 \times 10^{14}$ n/cm² sec time averaged
Spectrum: Roughly a fission reactor spectrum with a high energy tail
Availability: about 25 percent of the time at full flux (High Intensity Source), with possibility of operation at about 0.1 full flux (ZGS Booster II) during portions of the remaining time.

Based on the above characteristics, it is estimated that the annual fluence accumulation will be $\sim 1.5 \times 10^{21}$ n/cm² ($E > 0.1$ MeV).

B. FACILITIES

1. The Source

Some modifications of the IPNS Radiation Effects Source are suggested. (a) The tungsten moderators should be arranged in a grid similar to the fuel element arrangement in a conventional reactor. This will permit the adjustment of the experimental area next to the source to fit practical experimental needs. (b) Experimental space should be available at the bottom of the target to permit the study of radiation effects in a neutron spectrum which contains an exceptionally high portion of high energy neutrons (25-800 MeV).

2. Transfer Equipment

It is suggested that the cryogenic facility in the moderator be designed to allow removal of samples without warming up from the irradiation facility to remote testing cryostats for analysis by different methods. These methods

should include x-ray and low-energy neutron diffuse scattering (at IPNS) for identifying defect clusters.

3. Van de Graaff Accelerator

A charged particle accelerator (Van de Graaff ~ 2 MeV) should be available to the IPNS irradiation facility for two purposes: (a) for probing the defect state by Rutherford backscattering techniques, (b) for alternating e^- and neutron irradiations, which should allow determination of the effect of subthreshold collisions around defect cascades.

4. Thermal Neutron Irradiation Facility

An irradiation hole in the IPNS neutron scattering facility is suggested to be available for studies of thermal neutron damage.

5. Facilities for Mechanical Property Experiments at 0.2 to 0.5 T_m

Experimental facilities will be required to control, measure and record temperature, atmosphere, stress and strain.

6. Data Logging and Experimental Control Equipment

Appropriate electronic equipment for experimental control and data analysis including a microcomputer is required.

C. LOW TEMPERATURE ($T < 20$ K) IRRADIATION EFFECTS STUDIES

The IPNS is an attractive neutron source for pursuing radiation damage studies at low temperatures. This is a consequence of the low γ flux associated with the high neutron flux.

With modest refrigeration capacity bombardments at 40 K or lower can be maintained in fluxes on the order of 10^{14} n/cm² sec. This is a factor of about 20 over that now available. This high flux will permit a variety of experiments not now possible.

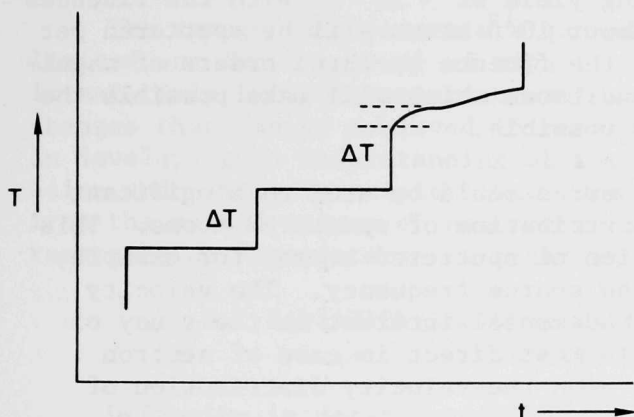
For example, one week bombardment will result in the sample attaining a defect concentration within 95% of the saturation limit in most materials including ordered alloys. This will permit the detailed study of the structure of displacement cascades in the region of high overlap. A variety of methods can be used for this study including resistivity, diffuse x-ray scattering, Rutherford backscattering and Mössbauer techniques provided the cryostat is designed to permit transfer at liquid helium temperatures. An important feature of this work is the determination of differences between the structure of the defect configuration in electron and neutron damage.

With the utilization of the lower fluxes available with injection by the ZGS Booster II the study of defect production from very low to very high fluences can be made. The effects of flux spectrum can also be studied by utilization of high energy neutrons which are dependent on vertical position.

The fact that the energy transferred to the sample by neutron collisions exceeds that due to γ interactions makes it possible to directly relate the energy transmitted by neutron collisions to the number of displacements by means of simultaneous calorimetric and resistometric measurements. Thus the comparison of defect production in a variety of metals can be made without utilization of uncertain elastic and inelastic cross section values.

It is also possible by the utilization of the short neutron pulses as a heat source to make a detailed study of the annealing behavior of irradiated metals. The technique is as follows. The heat input from the neutron pulse increases the sample temperature in steps as illustrated. This will cause defect migration and subsequent annihilation. The energy released by the annihilation will cause an additional increase in temperature. If the defects

annihilate with only a few jumps this increase in energy will be included in the neutron temperature step. Long range migration of the defects will result in a time delay permitting the separation of long range migration modes.



D. SUPERCONDUCTORS

The high flux of the IPNS and the ability to transfer samples to remote measuring systems at 4 K will make it possible to perform better basic as well as technologically interesting experiments on type II superconductors. In order to understand the elementary flux line-defect interaction it is necessary to measure the flux pinning as a function of temperature. This is presently not possible in low-temperature neutron facilities. Measuring the flux pinning in Nb (which saturates at $< 10^{18}$ n/cm²), after a dose that results in significant cascade overlap would help in understanding the decreased flux pinning efficiency of the cascades. Cryogenic irradiations of possible CTR magnet materials (NbTi, Nb₃Sn, V₃Ga) are presently limited to $< 10^{19}$ n/cm². The IPNS dose of 10^{20} n/cm²/week would make it possible to extend the fluence of irradiations of the superconductors as well as to irradiate small scale magnets. This latter type of irradiation would also give information on the insulators and potting material used in magnetic construction.

E. DAMAGE AT ELEVATED TEMPERATURE (20-300 K)

It will be possible to carry out rather careful damage rate measurements at temperatures at which damage production and annealing compete. Clustering of point defects at or near defect cascades as well as the influence of impurities could be studied, and by comparison with electron irradiation dose rate measurements, the importance of cascade damage on these effects could be studied.

In such experiments it is feasible to make use of the pulsed nature of the neutron source, since in principle it would be possible to separate production (within the pulses) and decay (between the pulses). However, this will be more difficult at low temperatures, and it is questionable whether this separation can be successfully done in metals; e.g., by resistivity measurements in the presently planned pulse structure. An increase of the neutron pulse (factor of ~ 100) and a corresponding decrease in pulse frequency would permit such experiments, however, as well as making it possible to separate the influences of instantaneous defect production and time/temperature on processes like vacancy and interstitial agglomeration.

F. SPUTTERING AND ELECTRON EMISSION

Sputtering studies must be made under carefully controlled conditions. Current measurements suggest a sputtering yield of $\sim 10^{-5}$. With the fluences available under these conditions only about 10^{12} atoms will be sputtered per cm^2 . It should be possible to increase the fluence by three orders of magnitude in IPNS with equally controlled conditions which will make possible the study of a variety of materials not now possible.

The pulsed nature of this neutron source could be used to significant advantage in determining the velocity distribution of sputtered atoms. This could be done by time-of-flight collection of sputtered atoms, for example, on a spinning rotor synchronized with the source frequency. The velocity distribution of sputtered atoms is of fundamental interest in the study of sputtering, and the relation to theory is most direct in case of neutron bombardment. The effect of temperature upon the velocity distribution of the sputtered atoms can be examined.

The relatively low gamma flux present during the operation of IPNS makes it possible to study secondary electron emission by neutron bombardment. This is an interesting new area previously not investigated.

G. MICROSTRUCTURAL EFFECTS

Radiation-produced changes in the microstructure of a metal include the formation of cavities (voids, bubbles), defect clusters, and dislocation structures (loops and networks). These structures are of importance due to their influence on such properties as swelling, electrical resistivity, strength, and fracture. These effects are generally produced in the range of operating conditions proposed for both fission and fusion reactors, that is, conditions of high neutron fluences at irradiation temperatures in the general range 0.2 to 0.6 T_m , where T_m is the melting point. These structural features result from the agglomeration of vacancies and interstitials, and are influenced by transmutation products, temperature, chemistry, displacement damage rate, and perhaps many other variables.

Irradiation of metal samples in the IPNS may be useful for the investigation of microstructural changes. The IPNS has the advantages of easy instrumentation, controllable irradiation temperature, and reasonably large experimental volume.

The pulsed nature of the IPNS neutron flux will affect the point defect concentrations in metals during irradiation. In the temperature range of interest, the concentration of free interstitials builds rapidly toward a steady state value, with the concentration of free vacancies climbing much more slowly to a steady state value. Steady state values are determined by the neutron flux (point defect generation rate) and the temperature-dependent diffusion coefficient for each defect. At the end of a neutron production pulse, the interstitial concentration will decay rapidly toward zero. Although time scales for the point defect build-up and decay are strongly temperature dependent, it is believed that interstitial decay, at least, will be rapid compared to the pulse repetition rate. This suggests that in IPNS irradiations different vacancy and interstitial concentrations will prevail from those in other currently available irradiation facilities. This unique feature of the facility should be considered.

The damage rate parameters quoted above show that the IPNS will be of limited use for the study of high fluence (~ 1 dpa) microstructural changes in materials for reactor structural applications. Study of microstructural changes that can be achieved in reasonable irradiation times will be helpful in developing an understanding of the mechanisms controlling defect agglomeration and cluster precipitation. The point defect concentrations that result from the pulsed nature of neutron irradiation could produce microstructural responses not encountered in other irradiation facilities.

H. RADIATION-ENHANCED DIFFUSION AND DIFFUSION-CONTROLLED METALLURGICAL REACTIONS

Diffusion in metals occurs by defect mechanisms. Diffusion is enhanced upon irradiation under conditions where an excess concentration of defects over the thermal equilibrium value is retained and where the temperature is high enough so that the defects are sufficiently mobile. Such diffusion enhancement can have significant effects on the structure and properties of alloys since it permits the approach toward equilibrium steady state configurations at low temperatures where atomic rearrangements would normally be limited because of low diffusivities.

Experimental observations have been made using steady-state neutron and charged particle irradiations of radiation-enhanced bulk diffusion and radiation-enhanced metallurgical reactions such as ordering, segregation, and precipitation. Kinetic models based upon various assumptions concerning annihilation mechanisms have been devised which permit a calculation of vacancy concentrations and diffusion enhancement as a function of vacancy and interstitial jump rates and production rates. The comparison of experiment and theory has not led to a definitive determination of the mechanism of radiation-enhanced diffusion, at least partly due to the limited range of defect production rates (instantaneous neutron fluxes) available to the experimenter. The availability of a neutron source providing instantaneous fluxes in the range of 10^{18-19} neutrons/cm² sec, as compared to 10^{12-13} neutrons/cm² sec for conventional experiments of the type discussed here, may permit further testing of the theories of radiation-enhanced diffusion.

Certain limitations of the IPNS in radiation-enhanced-diffusion research are also apparent. Each pulse of a high-flux radiation will be followed by a

relatively long wait time during which defect concentrations will decrease. The net effect of this somewhat complicated irradiation and annealing history is not clear. However, it can be said that exposure to the pulsed neutron environment of IPNS will constitute a vastly different set of irradiation conditions than has previously been experienced in radiation-enhanced diffusion research.

Depending upon temperature, and the distribution and density of sinks for point defects, differences between the quasi-steady state concentrations of vacancies and interstitials produced during pulsed neutron irradiations may be quite large. This effect may be useful in studying irradiation induced impurity/solute segregation in metals. Specifically, it may be possible to use such concentration differences to determine which of the point defect species is involved in segregating a given impurity/solute. For example, by varying temperatures and hence the relative concentrations of irradiation produced point defects, the extent of segregation may vary in a manner which allows the point defect species responsible to be determined. The usefulness of this technique is enhanced by the fact that the results may be easily compared to those obtained during steady-state neutron and heavy ion irradiations.

I. MECHANICAL PROPERTIES

The topics considered under mechanical properties include the following:

1. Radiation hardening
 - a. Increase in yield stress and flow stress
 - b. Increase in temperature dependence of yielding
 - c. Radiation-anneal hardening
2. Radiation Embrittlement
 - a. Dislocation channeling and radiation enhancement of plastic instability
 - b. Embrittlement of ductile-brittle metals; increase in ductile-brittle transition temperature
 - c. High-temperature grain-boundary embrittlement
3. Radiation enhanced creep and stress relaxation
4. Radiation enhanced fatigue

Fundamental research on the mechanisms of the above phenomena has involved in many cases neutron doses below, say, 10^{19} n/cm², which are also readily achievable in the IPNS.

Mechanical properties as influenced by radiation-produced defects depend on the barriers to slip dislocation motion provided by the defect clusters and by immobile dislocation structures. A matter of special interest is how the radiation-produced microstructural features characteristic of a pulsed high-energy neutron source like IPNS will affect the mechanical properties listed above. Theories of radiation hardening based on a dispersed barrier

model provide a possible method of correlating the density and size distribution of barriers to the observed radiation hardening. A study of such correlations for the IPNS radiation conditions may help to clarify the validity of radiation hardening models and thus increase the understanding of this subject.

Mechanical property studies on irradiated materials are sometimes limited by space consideration. However, adequate irradiation volumes appear to be incorporated in the IPNS radiation target design. Nevertheless, in the context of fracture research it is recognized that large-scale fracture mechanics samples cannot be accommodated. Gamma heating of thicker section samples is sometimes a problem, but this should not be the case for IPNS where gamma heating is quite low. The access to fairly large irradiation volumes should also facilitate the performance of in situ mechanical tests.

Radiation embrittlement is a complex and many-faceted subject. A wide range of research on radiation embrittlement can be carried out using irradiations in IPNS, although in some cases care will have to be exercised to select appropriate materials for study. Doses below 10^{20} or even 10^{19} neutrons/cm² are generally quite adequate for observing dislocation channels (and the resultant decrease in uniform and fracture strains) and for detecting increases in ductile-brittle transition temperature upon irradiation using either smooth-sample low-strain-rate tensile tests or notched-sample impact tests. The magnitude of high-temperature grain boundary embrittlement will depend largely on the amount of helium produced upon irradiation. Certainly, it will not be possible in IPNS to duplicate the very high fluence ($> 10^{21}$ n/cm²) effects used to study high-temperature grain boundary embrittlement in austenitic stainless steels and other cladding alloys. But higher purity materials will likely be susceptible to high-temperature grain boundary embrittlement upon irradiation in IPNS, particularly if a forward irradiation location is employed where the high-energy tail and therefore (n,α) reactions are enhanced.

The above discussion emphasizes the critical importance, vis-a-vis mechanical properties, of determining the neutron spectrum at the IPNS, particularly as regards the high-energy tail. Since transmutation products, especially helium, may have a severe embrittling effect, the neutron spectrum in IPNS and its influence on mechanical properties through transmutation should be carefully evaluated. Sufficient space will be available at IPNS to conduct irradiation creep and stress relaxation experiments. The creep data generated by in situ testing could be compared to those obtained by first irradiating samples and then post-irradiation-creep testing. The alloy constitution and its microstructure, temperature, flux, and stress conditions are important variables. IPNS would allow a more complete parametric evaluation of irradiation creep. Different fracture modes may well occur under irradiation creep conditions than are experienced under conventional conditions. Concerning the effect of the pulsed nature of the radiation in IPNS, some understanding may accrue from a comparison of results from this facility with those from LAMPF (with its different pulse structure).

J. CONSIDERATIONS ON THE RECOIL SPECTRUM

The neutron energy spectrum in the proposed IPNS radiation effects facility consists of two parts: a so-called nuclear evaporation component composed of neutrons in the 0.1 to 10 MeV range and a high energy component resulting from direct (intranuclear cascade) processes composed of neutrons of up to 800 MeV energy. The latter component is of considerable concern since it will produce not only the desired displacement of atoms in irradiated materials, but a much larger amount of transmutation than is customarily experienced in a fission spectrum. The effects of the transmutation products (H, He, and heavy elements as well) cannot be assessed with much reliability at this time, but may be of considerable importance in some kinds of experiments. The ratio of the two components in the neutron spectrum depends rather strongly on position within the W reflector, the high energy portion becoming most important in the region beneath the target. The resulting spatial differences offer opportunities for experimental separation of the effects of the two components as well as for experiments for which one or the other component is preferred. For this reason, ready access must be provided to all parts of the reflector, including the region directly below the target. Another approach to separating the effects of the high and low energy neutron components might be the use of a target other than the intended W one: the relative importance of the "hard" component would be greater from a light target such as C (graphite).

The close similarity of the neutron spectrum in the IPNS facility to that in the LAMPF beam stop should be noted. For many purposes there would be little to choose between the two. Thus, some of the problems of interpretation of experiments may be removed or alleviated by experiments run at LAMPF in the near future, long before the IPNS might be available. In both facilities, it will be necessary to have careful dosimetry, to establish the spectral shapes on a reliable basis before displacement damage experiments can be properly interpreted. Both also require a considerable expansion in the available cross section information, especially in the region from ~ 15 to ~ 50 MeV (below ~ 15 MeV the data are fairly complete; above ~ 50 MeV proton data can often be substituted). Cross sections are needed for neutron elastic and inelastic scattering, (n, xn) reactions, charged particle production reactions, etc., for each element to be irradiated in the facility. Without them, damage production data cannot be interpreted successfully. It may be that a program of cross section measurement could be established at either LAMPF or the IPNS to provide the needed data.

K. CONCLUSIONS

- (1) The IPNS is an attractive facility for basic radiation damage studies with neutrons. Such studies include low-temperature damage production and annealing, damage production at elevated temperature, mechanical properties, radiation-enhanced diffusion, microstructural studies and sputtering.
- (2) Specific limitations have been pointed out in the text.
- (3) The following features are of particular advantage for radiation effects studies:

- (a) The low level of gamma heating, especially as concerns low-temperature irradiation effects studies.
 - (b) The flux level is attractive.
 - (c) The radiation effects facility in IPNS is a dedicated source for radiation effects studies.
 - (d) The variation in neutron spectrum in different irradiation positions.
 - (e) Easy access to diagnostic tools and subsidiary irradiation facilities (e.g., the IPNS neutron scattering facility, van de Graaf accelerator, etc.).
 - (f) Large experimental volume.
 - (g) The possibility to utilize the pulsed nature of the beam.
 - (h) The low level of ionizing radiation may be important in some experiments.
- (4) The following points need further study:
- (a) More detailed analysis of the position-dependent neutron spectrum, in particular the influence of high-energy neutrons on the recoil spectrum.
 - (b) The influence of the pulse structure on high-temperature irradiation experiments may be important and could possibly be studied by comparison with LAMPF experiments.
 - (c) The dosimetry of the neutron beam.
 - (d) Possibilities of varying the pulse structure.

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The Neutron

$$m = 1.67495 \times 10^{-24} \text{ gm}, \quad \mu_n = 6.0311 \times 10^{-12} \text{ eV/gauss}$$

$$E = \frac{\hbar^2 k^2}{2m} = \frac{h^2}{2m\lambda^2} = hf = \frac{1}{2}mv^2 = \frac{1}{2}m\left(\frac{d}{t}\right)^2 = k_B T$$

$$E = 2.0717k^2 = \frac{81.787}{\lambda^2} = 4.1354f = 5.2276 \times 10^{-6} v^2$$

$$= 5.2276 \times 10^6 \frac{1}{(t/d)^2} = 0.086165T$$

$$E[\text{meV}], \lambda[\text{\AA}], f[\text{THz}], v[\text{m/sec}], k[\text{\AA}^{-1}], t/d\left[\frac{\mu\text{sec}}{\text{m}}\right], T[^\circ\text{K}]$$

$$\text{For a photon: } E(\text{meV}) = \frac{0.123975}{\lambda(\text{cm})}.$$

Conference and Workshop on Uses of Advanced Pulsed Neutron Sources,
Argonne National Laboratory, October 20-24, 1975